4th International Conference on Mathematical Modeling in Physical Sciences (IC-MSquare2015) IOP Publishing
Journal of Physics: Conference Series **633** (2015) 012063 doi:10.1088/1742-6596/633/1/012063

# Controlled Interaction of a Four-Level Quantum Emitter with a Plasmonic Nanostructure

**Emmanuel Paspalakis[1], Efthymios Kallos[1] and Vassilios Yannopapas[2]**

[1]Materials Science Department, School of Natural Sciences, University of Patras, Patras 265 04, Greece

[2]Department of Physics, National Technical University of Athens, Athens 157 80, Greece

E-mail: paspalak@upatras.gr, timaras@gmail.com, vyannop@mail.ntua.gr

**Abstract.** We present results on the controlled dynamics of an externally driven four-level quantum emitter coupled to a plasmonic metamaterial, specifically a periodic two-dimensional array of metal-coated dielectric nanospheres. For the study of the system's dynamics, we combine the density matrix approach for the quantum emitter with *ab initio* electromagnetic calculations for the plasmonic nanostructure. We then present results for the time evolution of the populations of the different levels of the quantum emitter in both the presence and the absence of the plasmonic nanostructure. The dependence of the population dynamics to different distances of the quantum emitter from the plasmonic nanostructure is also studied.

## 1. Introduction

Recently, there is increasing interest in the study of the interaction of quantum emitters (such as atoms, molecules and semiconductor quantum dots) with plasmonic nanostructures [1]. The large fields and the strong light confinement associated with the plasmonic resonances enable strong interaction between the electromagnetic field and the quantum emitters near plasmonic nanostructures. Also, the quantum emitter can be used for the controlled optical response of the hybrid quantum - plasmonic system.

A quantum emitter that has attracted particular attention is modeled by a four-level quantum system with two V-type transitions [2, 3, 4, 5, 6, 7, 8]. In the quantum system one V-type transition is influenced by the interaction with surface plasmons while the other V-type transition interacts with free space vacuum [see Fig. 1(a)]. This system has led to several interesting quantum coherence and interference phenomena, including optical transparency and slow light [2], transient gain without inversion [3], strongly modified Kerr nonlinearity [4], phase-dependent absorption and dispersion [5], strongly modified spontaneous emission spectrum [6, 7] and coherent population trapping [8].

In this work, we present new theoretical results on the controlled dynamics of the four-level quantum emitter coupled to a plasmonic metamaterial, namely a periodic two-dimensional array of metal-coated dielectric nanospheres. An external electromagnetic field is applied to the system and is used for the control of the dynamics of the quantum system. For the study of the system's dynamics, we combine the density matrix approach for the quantum emitter with *ab initio* electromagnetic calculations for the plasmonic nanostructure. We then present results for the time evolution of the population (occupation probability) of the different levels of the

quantum emitter in both the presence and the absence of the plasmonic nanostructure. Also, in the presence of the plasmonic nanostructure, we consider the influence of different distances of the quantum emitter from the plasmonic nanostructure to the population dynamics.

## 2. Theoretical Model

The quantum system of interest is shown in Fig. 1(a). We consider a four-level system with two closely lying upper states $|2\rangle$ and $|3\rangle$, and two lower states $|0\rangle$ and $|1\rangle$. The quantum system is located in vacuum at distance $D$ from the surface of the plasmonic nanostructure. We take states $|2\rangle$ and $|3\rangle$ to characterize two Zeeman sublevels. The dipole moment operator is taken as $\vec{\mu} = \mu^{'}(|2\rangle\langle 0|\hat{\varepsilon}_{-} + |3\rangle\langle 0|\hat{\varepsilon}_{+}) + \mu(|2\rangle\langle 1|\hat{\varepsilon}_{-} + |3\rangle\langle 1|\hat{\varepsilon}_{+}) + \text{H.c.}$, where $\hat{\varepsilon}_{\pm} = (\mathbf{e}_z \pm i\mathbf{e}_x)/\sqrt{2}$, describe the right ($\hat{\varepsilon}_{+}$) and left-rotating ($\hat{\varepsilon}_{-}$) unit vectors; and $\mu$, $\mu^{'}$ are taken to be real.

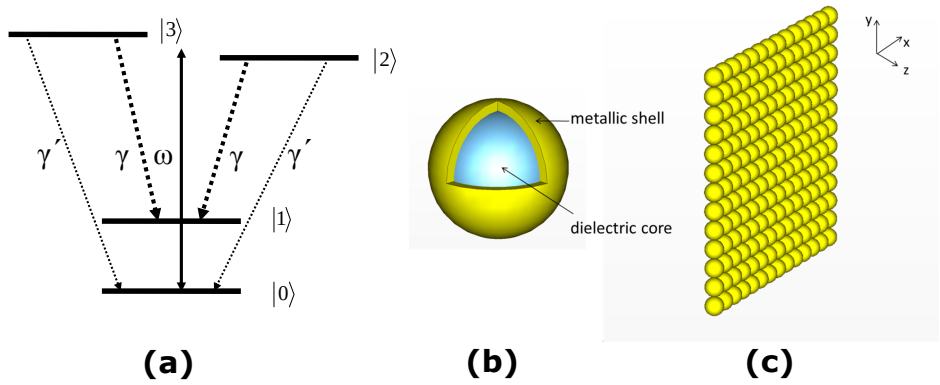


**Figure 1.** (a) The quantum system under study is a four-level system where two upper states $|2\rangle$ and $|3\rangle$ decay with spontaneous emission to the two lower states $|0\rangle$ and $|1\rangle$. The system interacts with an electromagnetic field that couples state $|0\rangle$ with states $|2\rangle$ and $|3\rangle$. (b) A metal-coated dielectric nanosphere and (c) the two-dimensional array of metal-coated dielectric nanospheres used in this work.

The quantum system interacts with a linearly polarized continuous wave laser field, with electric field $\vec{E}(t) = \hat{z}E_0\cos(\omega t)$, where $E_0$ is the electric field amplitude and $\omega$ the angular frequency of the electric field. The laser field couples state $|0\rangle$ with states $|2\rangle$ and $|3\rangle$.

Both excited states $|2\rangle$ and $|3\rangle$ decay spontaneously to state $|0\rangle$ with decay rate $2\gamma^{'}$ and to state $|1\rangle$ with decay rate $2\gamma$. We assume that the transitions $|2\rangle$, $|3\rangle$ to $|1\rangle$ lie within the surface-plasmon bands of the plasmonic nanostructure, whereas the transitions $|2\rangle$, $|3\rangle$ to $|0\rangle$ are spectrally far from the surface-plasmon bands and are not influenced by the plasmonic nanostructure.

The Hamiltonian that describes the interaction of the electromagnetic field with the quantum system, in the dipole and rotating wave approximations, is given by

$$\begin{aligned} H &= \hbar\left(-\delta - \frac{\omega_{32}}{2}\right)|2\rangle\langle 2| + \hbar\left(-\delta + \frac{\omega_{32}}{2}\right)|3\rangle\langle 3| \\ &- \frac{\hbar\Omega}{2}\left(|0\rangle\langle 2| + |0\rangle\langle 3| + H.c.\right). \end{aligned} \tag{1}$$

Here, $\delta = \omega - \tilde{\omega}$ is the detuning from resonance with the average transition energies of states $|2\rangle$ and $|3\rangle$ from state $|0\rangle$, with $\tilde{\omega} = (\omega_3 + \omega_2)/2 - \omega_0$, $\omega_{32} = (\omega_3 - \omega_2)/2$, and $\Omega$ is the Rabi frequency defined as $\Omega = \mu^{'}E_0/(\sqrt{2}\hbar)$. Also, $\hbar\omega_n$ with $n = 0 - 3$, is the energy of state $|n\rangle$.

Using the Hamiltonian of Eq. (1) we obtain the following equations for the density matrix elements of the system, assuming a Markovian response:

$$\begin{aligned}
\dot{\rho}_{00}(t) &= 2\gamma^{'}\left[\rho_{22}(t)+\rho_{33}(t)\right]-i\frac{\Omega}{2}\left[\rho_{02}(t)-\rho_{20}(t)\right] \\
&- i\frac{\Omega}{2}\left[\rho_{03}(t)-\rho_{30}(t)\right], \qquad (2)\\
\dot{\rho}_{22}(t) &= -2(\gamma+\gamma^{'})\rho_{22}(t)+i\frac{\Omega}{2}\left[\rho_{02}(t)-\rho_{20}(t)\right] \\
&- \kappa\left[\rho_{23}(t)+\rho_{32}(t)\right], \qquad (3)\\
\dot{\rho}_{33}(t) &= -2(\gamma+\gamma^{'})\rho_{33}(t)+i\frac{\Omega}{2}\left[\rho_{03}(t)-\rho_{30}(t)\right] \\
&- \kappa\left[\rho_{23}(t)+\rho_{32}(t)\right], \qquad (4)\\
\dot{\rho}_{20}(t) &= (i\delta+i\frac{\omega_{32}}{2}-\gamma-\gamma^{'})\rho_{20}(t)-i\frac{\Omega}{2}\rho_{22}(t) \\
&- i\frac{\Omega}{2}\rho_{23}(t)+i\frac{\Omega}{2}\rho_{00}(t)-\kappa\rho_{30}(t), \qquad (5)\\
\dot{\rho}_{30}(t) &= (i\delta-i\frac{\omega_{32}}{2}-\gamma-\gamma^{'})\rho_{30}(t)-i\frac{\Omega}{2}\rho_{33}(t) \\
&- i\frac{\Omega}{2}\rho_{32}(t)+i\frac{\Omega}{2}\rho_{00}(t)-\kappa\rho_{20}(t), \qquad (6)\\
\dot{\rho}_{23}(t) &= (i\omega_{32}-2\gamma-2\gamma^{'})\rho_{23}(t)+i\frac{\Omega}{2}\rho_{03}(t)-i\frac{\Omega}{2}\rho_{20}(t) \\
&- \kappa\left[\rho_{22}(t)+\rho_{33}(t)\right], \qquad (7)
\end{aligned}$$

with $\rho_{00}(t)+\rho_{11}(t)+\rho_{22}(t)+\rho_{33}(t)=1$ and $\rho_{nm}(t)=\rho^{*}_{mn}(t)$. Here, $\kappa$ is the coupling coefficient between states $|2\rangle$ and $|3\rangle$ due to spontaneous emission in a modified anisotropic vacuum [9]. This term is responsible for the appearance of quantum interference in spontaneous emission [10].

The values of $\gamma$ and $\kappa$ are obtained by [9, 11, 12]

$$\gamma = \frac{\mu_0\mu^2\bar{\omega}^2}{\hbar}\hat{\varepsilon}_{-}\cdot\mathrm{Im}\mathbf{G}(\mathbf{r},\mathbf{r};\bar{\omega})\cdot\hat{\varepsilon}_{+}=\frac{1}{2}\left(\Gamma_{\perp}+\Gamma_{\parallel}\right), \qquad (8)$$

$$\kappa = \frac{\mu_0\mu^2\bar{\omega}^2}{\hbar}\hat{\varepsilon}_{+}\cdot\mathrm{Im}\mathbf{G}(\mathbf{r},\mathbf{r};\bar{\omega})\cdot\hat{\varepsilon}_{+}=\frac{1}{2}\left(\Gamma_{\perp}-\Gamma_{\parallel}\right). \qquad (9)$$

Here, $\mathbf{G}(\mathbf{r},\mathbf{r};\omega)$ is the dyadic electromagnetic Green's tensor, where $\mathbf{r}$ refers to the position of the quantum emitter, and $\mu_0$ is the permeability of vacuum. Also, $\bar{\omega}=(\omega_3+\omega_2)/2-\omega_1$. In addition, we define the spontaneous emission rates normal and parallel to the surface as $\Gamma_{\perp,\parallel}=\mu_0\mu^2\bar{\omega}^2\mathrm{Im}[G_{\perp,\parallel}(\mathbf{r},\mathbf{r};\bar{\omega})]/\hbar$, where $G_{\perp}(\mathbf{r},\mathbf{r};\bar{\omega})=G_{zz}(\mathbf{r},\mathbf{r};\bar{\omega})$, $G_{\parallel}(\mathbf{r},\mathbf{r};\bar{\omega})=G_{xx}(\mathbf{r},\mathbf{r};\bar{\omega})$ denote components of the electromagnetic Green's tensor where the symbol $\perp$ ($\parallel$) refers to a dipole oriented normal - along the $z$-axis (parallel - along the $x$-axis) to the surface of the nanostructure.

The plasmonic nanostructure considered in this study is a two-dimensional array of touching silver-coated silica nanospheres [see Fig. 1(b) and (c)]. The dielectric function of the shell is provided by a Drude-type electric permittivity given by

$$\epsilon(\omega)=1-\frac{\omega_p^2}{\omega(\omega+i/\tau)}, \qquad (10)$$

where $\omega_p$ is the bulk plasma frequency and $\tau$ the relaxation time of the conduction-band electrons of the metal. The dielectric constant of $SiO_2$ is taken to be $\epsilon=2.1$. In the calculations we have

**Table 1.** The values of $\Gamma_\perp$ and $\Gamma_\parallel$ for different distances $D$ from the surface of the plasmonic nanostructure for $\hbar\bar{\omega} = 2.4$ eV. $\Gamma_0$ is the spontaneous decay rate in free space and in our work we take it $\Gamma_0 = 10^9$ ns$^{-1}$.

| $D$ $(nm)$ | $\Gamma_\perp$ $(\Gamma_0)$ | $\Gamma_\parallel$ $(\Gamma_0)$ |
|---|---|---|
| 10.4 | 27.081 | 0.105 |
| 20.8 | 6.417 | 0.038 |
| 31.2 | 1.774 | 0.021 |
| 41.6 | 0.559 | 0.021 |
| 52 | 0.196 | 0.026 |

taken $\tau^{-1} = 0.1\omega_p$ and $\hbar\omega_p = 3.8$ eV corresponding to silver. The lattice constant of the square lattice is $a = 104$ nm and the sphere radius $S = 52$ nm with core radius $S_c = 36.4$ nm.

## 3. Numerical Results

The corresponding electromagnetic Green's tensor which provides the corresponding spontaneous emission rates $\Gamma_\perp$ and $\Gamma_\parallel$ is given by [12, 13, 14]

$$G^{EE}_{ii'}(\mathbf{r},\mathbf{r}';\omega) = g^{EE}_{ii'}(\mathbf{r},\mathbf{r}';\omega) - \frac{i}{8\pi^2}\int\int_{SBZ} d^2\mathbf{k}_\parallel \sum_{\mathbf{g}} \frac{1}{c^2 K^+_{\mathbf{g};z}} \times v_{\mathbf{g}\mathbf{k}_\parallel;i}(\mathbf{r}) \exp(-\mathrm{i}\mathbf{K}^+_{\mathbf{g}}\cdot\mathbf{r})\hat{\mathbf{e}}_{i'}(\mathbf{K}^+_{\mathbf{g}}), \tag{11}$$

with

$$v_{\mathbf{g}\mathbf{k}_\parallel;i}(\mathbf{r}) = \sum_{\mathbf{g}'} R_{\mathbf{g}';\mathbf{g}}(\omega,\mathbf{k}_\parallel)\exp(-\mathrm{i}\mathbf{K}^-_{\mathbf{g}'}\cdot\mathbf{r})\hat{\mathbf{e}}_i(\mathbf{K}^-_{\mathbf{g}'}), \tag{12}$$

and

$$\mathbf{K}^\pm_{\mathbf{g}} = (\mathbf{k}_\parallel + \mathbf{g},\ \pm[q^2 - \left(\mathbf{k}_\parallel + \mathbf{g}\right)^2]^{1/2}). \tag{13}$$

The vectors $\mathbf{g}$ denote the reciprocal-lattice vectors corresponding to the 2D periodic lattice of the plane of scatterers and $\mathbf{k}_\parallel$ is the reduced wavevector which lies within the surface Brillouin zone associated with the reciprocal lattice [15]. When $q^2 = \omega^2/c^2 < (\mathbf{k}_\parallel + \mathbf{g})^2$, $\mathbf{K}^\pm_{\mathbf{g}}$ defines an evanescent wave. The term $g^{EE}_{ii'}(\mathbf{r},\mathbf{r}';\omega)$ of Eq. (11) is the free-space Green's tensor and $\hat{\mathbf{e}}_i(\mathbf{K}^\pm_{\mathbf{g}})$ the polar unit vector normal to $\mathbf{K}^\pm_{\mathbf{g}}$. $R_{\mathbf{g}';\mathbf{g}}(\omega,\mathbf{k}_\parallel)$ is the reflection matrix which provides the sum (over $\mathbf{g}$'s) of reflected beams generated by the incidence of plane wave from the left of the plane of scatterers [15]. Also, in Eq. (11), the terms corresponding to $s$-polarized waves (those containing components with the azimuthial unit vector $\hat{\mathbf{e}}_i(\mathbf{K}^\pm_{\mathbf{g}})$ normal to $\mathbf{K}^\pm_{\mathbf{g}}$) have small contribution to the decay rates and have been, therefore, neglected.

We use the above methodology and calculate the spontaneous emission rates normal and parallel to the surface of the plasmonic nanostructure for different distances of the quantum emitter from the plasmonic nanostructure. The results of the calculations are shown in Table 1.

We assume that the quantum system is initially in state $|0\rangle$. This means $\rho_{00}(0) = 1$ and for all other elements $\rho_{nm}(0) = 0$. We are interested in the time evolution of the populations $\rho_{nn}(t)$, with $n = 0, 2, 3$, at $\delta = 0$ $(\omega = \tilde{\omega})$, because, at this frequency, coherent population trapping may occur in states $|0\rangle$, $|2\rangle$ and $|3\rangle$ if $\kappa = \gamma$ and $\gamma' = 0$ [8, 16, 17]. Here, $\kappa$ and $\gamma$ are never exactly equal to each other (see the values of Table 1); however, we expect an interesting behavior of the population dynamics at this frequency if $\gamma' = 0$. Therefore, we will present results obtained from a numerical solution of the density matrix equations (2)-(7) using a fourth-order Runge-Kutta method.

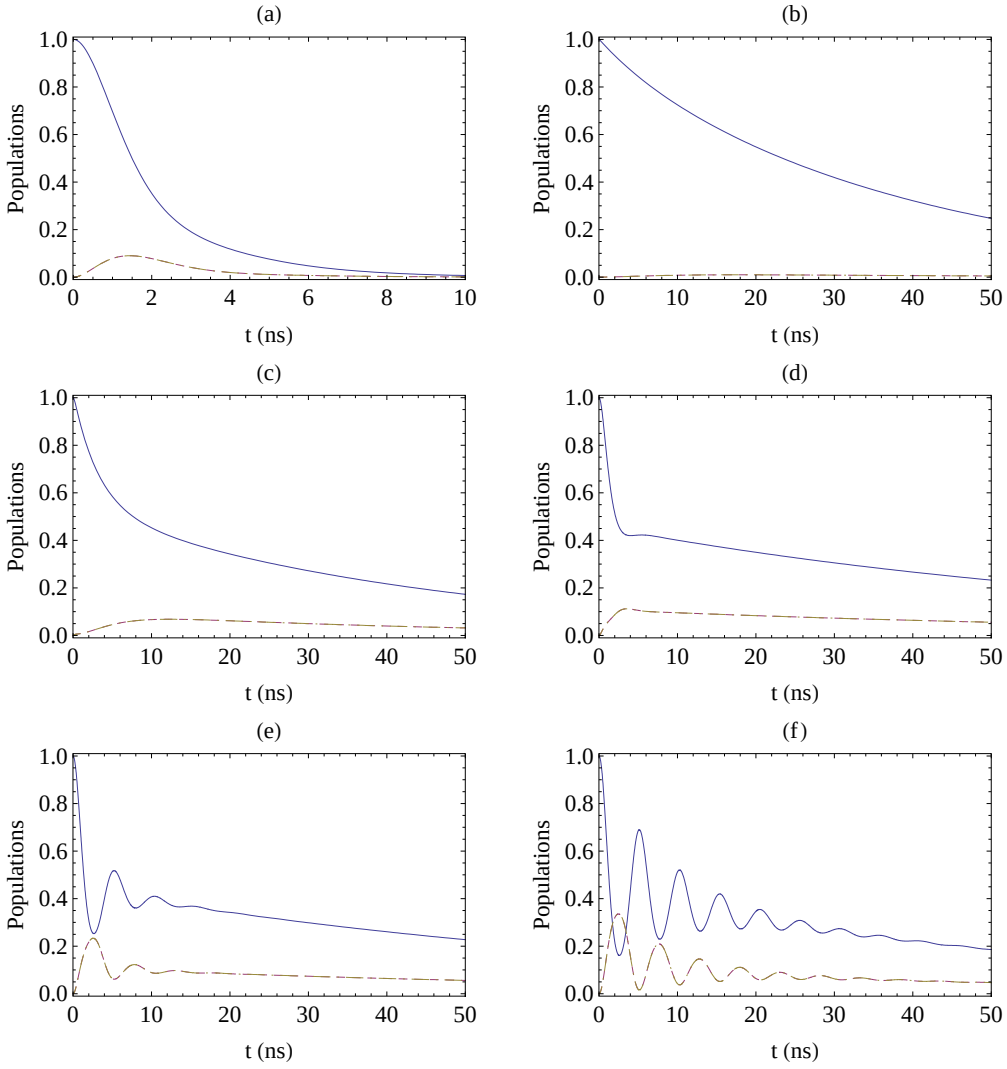


**Figure 2.** The time evolution of the population $\rho_{nn}(t)$ in states $|n\rangle$. With solid curve we present the population in state $|0\rangle$, with dashed curve the population in state $|2\rangle$ and with dot-dashed curve the population in state $|3\rangle$. Plot (a) shows results in the absence of the plasmonic nanostructure, while the other plots depict results in the presence of the plasmonic nanostructure. We take $\hbar\bar{\omega} = 2.4$ eV, $\Omega = 1$ ns$^{-1}$, $\omega_{32} = 2$ ns$^{-1}$ and $\gamma^{'} = 0$. In (b) $D = 10.4$ nm, (c) $D = 20.8$ nm, (d) $D = 31.2$ nm, (e) $D = 41.6$ nm and (f) $D = 52$ nm.

In Fig. 2 we present the population $\rho_{nn}(t)$, with $n = 0, 2, 3$ as a function of time for $\gamma^{'} = 0$. We note that $\rho_{22}(t) = \rho_{33}(t)$. When the quantum system is placed in vacuum [Fig. 2(a)], i.e. in the absence of the plasmonic nanostructure, a transient weak excitation of the population to states $|2\rangle$ and $|3\rangle$ occurs and the whole population is transfer by spontaneous emission to state $|1\rangle$ in about 10 ns. In the presence of the plasmonic nanostructure [Figs. 2(b)-(f)] the evolution of the population is different and depends on the distance of the quantum emitter from the plasmonic nanostructure. For distances close to the plasmonic nanostructure, such those shown in Figs. 2(b) and (c), the population remains in states $|0\rangle$ (mainly), $|2\rangle$ and $|3\rangle$ for much longer times. For even longer distances, Fig. 2(d), a weak oscillation is obtained together with the slow population decay. Similar behavior is also found for distances close or equal to the nanoparticle

radius, Figs. 2(e) and (f), with the addition that in this case the populations exhibit stronger damped oscillations. We note that similar behavior to the last figure is obtained for distances that are within a few nanoparticle radii. For much larger distances the influence of the plasmonic nanostructure becomes very weak, with no signature of the plasmonic nanostructure.

## 4. Summary

We have studied the controlled dynamics of the four-level quantum emitter coupled to a plasmonic metamaterial. The plasmonic system we examined is a periodic two-dimensional array of silver-coated silica nanospheres. An external electromagnetic field is applied to the system and is used for the control of the dynamics of the quantum emitter. For the study of the system's dynamics, we combined the density matrix approach for the quantum emitter with *ab initio* electromagnetic calculations for the plasmonic nanostructure. We then presented results for the time evolution of the population of the different levels of the quantum emitter in both the presence and the absence of the plasmonic nanostructure. We found that the population evolution depends strongly on the distance between the quantum emitter and the plasmonic nanostructure.

## Acknowledgments

This work was implemented within the framework of the Action Supporting Postdoctoral Researchers of the Operational Program Education and Lifelong Learning (Actions Beneficiary: General Secretariat for Research and Technology), and was co-financed by the European Social Fund and the Greek State (Program Nanokallos PE3_26).

## References

[1] Tame M S, McEnery K R, Ozdemir S K, Lee J, Maier S A and Kim M S 2013 *Nature Phys.* **9** 329.
[2] Evangelou S, Yannopapas V and Paspalakis E 2012 *Phys. Rev. A* **86** 053811.
[3] Evangelou S, Yannopapas V and Paspalakis E 2014 *Opt. Commun.* **314** 36.
[4] Evangelou S, Yannopapas V and Paspalakis E 2014 *J. Mod. Opt.* **61** 1458.
[5] Paspalakis E, Evangelou S, Yannopapas V and Terzis A F 2013 *Phys. Rev. A* **88** 053832.
[6] Evangelou S, Yannopapas V and Paspalakis E 2011 *Phys. Rev. A* **83** 023819.
[7] Gu Y, Wang L, Ren P, Zhang J-X, Zhang T-C, Martin O J F and Gong Q-H 2012 *Nano Lett.* **12** 2488.
[8] Gu Y, Wang L, Ren P, Zhang J-X, Zhang T-C, Xu J-P, Zhu S-Y and Gong Q-H 2012 *Plasmonics* **7** 33.
[9] Agarwal G S 2000 *Phys. Rev. Lett.* **84** 5500.
[10] Kiffner M, Macovei M, Evers J and Keitel C H 2010 *Prog. Opt.* **55** 85.
[11] Yang Y-P, Xu J-P, Chen H and Zhu S-Y 2008 *Phys. Rev. Lett.* **100** 043601.
[12] Yannopapas V, Paspalakis E and Vitanov N V 2009 *Phys. Rev. Lett.* **103** 063602.
[13] Sainidou R, Stefanou N and Modinos A 2004 *Phys. Rev. B* **69** 064301.
[14] Yannopapas V and Vitanov N V 2007 *Phys. Rev. B* **75** 115124.
[15] Stefanou N, Yannopapas V and Modinos A 1998 *Comput. Phys. Commun.* **113**, 49;
2000 *Comput. Phys. Commun.* **132** 189.
[16] Zhu S Y and Scully M O 1996 *Phys. Rev. Lett.* **76** 388.
[17] Paspalakis E and Knight P L 1998 *Phys. Rev. Lett.* **81** 293.