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Spontaneous radiation of a two-level atom into multipole modes of a plasmonic nanoparticle☆

E.S. Andrianov [a,b,*], A.A. Pukhov [a,b,c], A.P. Vinogradov [a,b,c], A.V. Dorofeenko [a,b,c], A.A. Lisyansky [d]

[a] All-Russia Research Institute of Automatics, 22 Sushchevskaya, Moscow 127055, Russia
[b] Moscow Institute of Physics and Technology, 9 Institutskiy per., Dolgoprudniy 141700, Moscow Reg., Russia
[c] Institute for Theoretical and Applied Electromagnetics RAS, Moscow, Russia
[d] Department of Physics, Queens College of the City University of New York, Queens, NY 11367, USA

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Abstract

We consider the relaxation of an excited two-level system (TLS) positioned near a spherical plasmonic nanoparticle (NP). The transition frequency of the TLS is assumed to coincide with the frequency of the condensation point of NP plasmonic resonances. We show that the relaxation of the TLS excitation is a two-step process. Following an initial exponential decay, the TLS breaks in to Rabi oscillations. Depending upon the distance between the TLS and NP, the probability of the TLS being in the excited state exhibits either chaotic or nearly regular oscillations. In the latter case, the eigenfrequency of the TLS-NP system coincides with one of NP multipole modes.

Keywords: Plasmonics; Metamaterials; Spontaneous emission; Two-level system

1. Introduction

The problem of the giant decrease of the radiative relaxation time of atoms near metallic NPs has attracted considerable interest in the last decade due to explosive growth of nanoplasmonics [1–6]. In the case of interaction with countable number of high multipole modes the relaxation is much more complicated than in simple cases of dipole moment relaxation, which may be the exponential fall-off of a TLS excitation into a continuum of modes [7] or the Rabi oscillations when the TLS energy is transferred into a single resonant mode [8]. It has been shown recently [9], that the rate of spontaneous exponential nonradiative decay in a TLS, which is in resonance with the dipole mode of a lossy plasmonic sphere, increases by several orders of magnitude thanks to accounting of higher non-resonant multipole modes. This qualitatively agrees with ideas of Ref. [10] that though the contribution of higher multipoles to photon radiation is smaller than that of the dipole, Joule dissipation of higher multipoles is significantly greater than the dissipation of the dipole. The increase of dissipation leads to a decrease of the life-time of the emitter.

In the case of a metallic NP, the energy can be transferred into an infinite but countable set of modes which

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* Corresponding author at: All-Russia Research Institute of Automatics, 22 Sushchevskaya, Moscow 127055, Russia.
Tel.: +7 9853632369.
E-mail address: evgeniy.andrianov@phystech.edu
(E.S. Andrianov).

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spectrum has a condensation point. It is not intuitively clear how a TLS, which is in resonance with the NP condensation point would decay. One might expect that the relaxation of this system due to coupling to an infinite number of modes is similar to the case of coupling to a continuum of modes. On the other hand, the relaxation into the condensation point may resemble the relaxation into a single resonant mode.

In this paper, we study the spontaneous relaxation of a TLS, with a transition frequency in resonance with the frequency of the condensation point of higher multipole resonances. We show that the relaxation of the TLS into a condensation point of the NP plasmonic spectrum is quite unusual. It occurs in three stages. In the initial stage, it has the exponential character due to the existence of infinite albeit countable number of modes to which it couples. This is similar to the case of continuum of modes. However, the system relaxes not into the ground state but toward a quasi-stationary state. Then, in the second stage, the exponential decay transforms into the Rabi oscillations. In the latter stage, the probability of the TLS to be in the excited state, exhibits either chaotic or nearly regular oscillations depending upon the distance between the TLS and NP. Finally, at the third stage, the relaxation is again exponential due to Joule losses in metal.

The problem of atomic relaxation near a metallic NP requires a quantum description of the field and radiating system [11–14]. The simplest approach using the Fermi Golden Rule does not produce the radiation spectrum unless an additional assumption is made about the Lorentzian line shape, which arises from the interaction of the TLS with continuum of modes [7]. To obtain the decay law from the first principles without additional assumptions we use the Weisskopf–Wigner [8,14].

2. Time evolution in the limiting cases of continuum of modes and single mode

The general analysis of a TLS interacting with cavity modes is given in Ref. [7]. Let us consider a set $|k\rangle$ of resonator modes and excited $|e\rangle$ and ground $|g\rangle$ states of the TLS. In order to describe the relaxation of the excited state of the TLS, we assume that each resonator mode can interact with a continuum of modes kk' of some reservoir. If the corresponding kk' modes are phonons in metal, the interaction reduces to the Joule losses in the NP. So we may take the Hamiltonian of system in the form

$$\hat{H} = \hbar\omega_{\mathrm{TLS}}\hat{\sigma}^{\dagger}\hat{\sigma} + \hbar\sum_k \omega_k \hat{a}_k^{\dagger}\hat{a}_k + \hbar\sum_k \gamma_k\left(\hat{a}_k^{\dagger}\hat{\sigma} + \hat{\sigma}^{\dagger}\hat{a}_k\right) + \hbar\sum_{k'}\omega_{k'}\hat{b}_{k'}^{\dagger}\hat{b}_{k'} + \hbar\sum_{kk'}\Gamma_{kk'}\left(\hat{a}_k^{\dagger}\hat{b}_{kk'} + \hat{b}_{kk'}^{\dagger}\hat{a}_k\right). \tag{1}$$

In the Hamiltonian (1), the first term corresponds to the TLS energy (ω_{TLS} is the TLS transition frequency, $\hat{\sigma}$ is the transition operator between ground and excited states of the TLS), the second term describes the energy of each multipole (ω_k is the resonance frequency of the kth multipole mode of the NP, $\hat{a}_k$ is the plasmon annihilation operator), the third term corresponds to the interaction of the TLS and the kth multipole mode (γ_k is the interaction constant), the fourth term corresponds to the energy of the thermal reservoir (ω_k is the eigenfrequency of k'th multipole mode of the NP, $\hat{b}_{k'}$ is the annihilation operator of this mode), and the last term describes the interaction between kth and k'th multipole modes of the thermal bath ($\Gamma_{kk'}$ is the interaction constant).

Let us expand the wave function of the system "atom + resonator modes + reservoir modes" over the stationary states in absence of interactions

$$\begin{aligned}\Psi(t) = {} & A(t)\exp(-i\omega_{\mathrm{TLS}}t)\,|e, 0, 0\rangle \\ & + \sum_k B_k(t)\exp(-i\omega_k t)\,|g, 1_k, 0\rangle \\ & + \sum_{k,k'} C_{kk'}(t)\exp(-i\omega_{kk'}t)\,|g, 0, 1_{kk'}\rangle,\end{aligned} \tag{2}$$

where $|e, 0, 0\rangle$ denotes the state in which only the atom is excited, $|g, 1_k, 0\rangle$ is the state in which only the kth mode of the resonator is excited, and $|g, 0, 1_{kk'}\rangle$ denotes the state in which only k'th wall mode of the kth mode of the resonator is excited.

Using the Schrödinger equation with Hamiltonian (1) and expansion (2) we obtain the system of equations

$$i\dot{A}(t) = \sum_k \gamma_k^* B_k(t)\exp(-i(\omega_k - \omega_{\mathrm{TLS}})t), \tag{3}$$

$$\begin{aligned} i\dot{B}_k(t) = {} & \gamma_k A(t)\exp(i(\omega_k - \omega_{TLS})t) \\ & + \sum_{k'} \Gamma_{kk'}^* C_{kk'}(t)\exp(-i(\omega_{kk'} - \omega_k)t),\end{aligned} \tag{4}$$

$$i\dot{C}_{kk'}(t) = \Gamma_{kk'} B_k(t)\exp(i(\omega_{kk'} - \omega_k)t). \tag{5}$$

Taking into account the initial conditions $A(0) = 1$, $B_k(0) = 0$, $C_{kk'}(0) = 0$ and using the Fourier transformation

$$\alpha(q) = \frac{1}{2\pi}\int_0^{\infty} A(t)\exp(iqt)dt, \tag{6}$$