Fluorescence power of an atom near polarizable particles

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Abstract. We present a semi classical model for studying the fluorescent power radiated by an atom placed at the interstices between polarizable particles with dimensions at the nanometer scale irradiated by a laser with frequency close to resonance. The atom is represented by a two-level quantum system and for the electromagnetic response of the surrounding particles a model based on their multipolar polarizability is used. Results are shown for the case of an atom in between two spherical particles forming a cluster, giving the fluorescent power of the atom as a function of the distance to the spheres and also of the laser frequency and intensity.

1.1. Introduction

The radiation emitted by an atom is strongly affected by the presence of polarizable particles. The induced electric charge distribution over these particles affect the local field acting upon the atom and a modification on lifetime of excited states is observed \cite{1,2}. This effect is relevant to radiation emission processes such as SERS \cite{3,4} and fluorescence \cite{5,6}. In a previous work we have shown that for a molecule near a metallic surface, fluorescence due to spontaneous emission may be enhanced by up to sixth orders of magnitude \cite{7}.

For the case of atoms surrounded by metallic particles irradiated by a laser, there is an induced charge distribution over the particles, producing an increase of the local electromagnetic intensity and a corresponding increase in the transition rate and radiated power (fluorescence enhancement effect). This effect is more significant as there is a reduction in dimensions of the interstice where the atom is placed. In addition, non-radiative energy transfer to the particles helps to reduce this rate (quenching fluorescence), both effects being competitive \cite{5}.

In this work we use a semi classical approach based on normal modes theory for particles arrays \cite{8,9}, and we calculate the power radiated by an atom close to mesoscopic objects such as nanometrical spheres under the influence of a laser field.

Results for the fluorescence radiated power are shown for the case of an atom placed in between two dielectric spheres, by varying the distance to the spheres and the laser frequency.
2. Fluorescence of an atom close to spherical particles
We consider an atom placed close to polarizable particles in the presence of a laser external field of frequency \( \omega \). From the solution to the optical Bloch equations [11], it can be obtained the steady state power radiated by an isolated two level atom. It can be written in terms of the Rabi frequency \( \eta \), the transition frequency \( \omega_o \) between levels and the spontaneous decay rate \( \gamma \) from the excited to the ground state. This decay rate is strongly influenced by its environment [12], [13], and the Rabi frequency depends on the local electric field at the site where the atom is placed.

The combination of fluorescent enhancement and fluorescence quenching as two competitive effects, leads to the following expression for the fluorescent power normalized to vacuum (isolated atom):

\[
P_{fluores} = G(\omega)^4 \cdot M(\omega) \left[ 1 + \delta^2 + \frac{1}{2} \left( \frac{\eta_o}{\gamma_o} \right)^2 \right] \left[ M(\omega)^2 + \delta^2 + \frac{1}{2} G^2 \left( \frac{\eta_o}{\gamma_o} \right)^2 \right],
\]

Here \( G(\omega) \) is the amplification factor in the magnitude of the local electric field acting upon the atom, \( \delta \) is the detuning factor defined as \( \delta = \left( \omega - \omega_o \right) / \gamma_o \). The frequency dependent factor \( M(\omega) \) represents the modification in the transition rate due to the ohmic losses increase when an atom is placed near metallic nanoparticles.

3. Numerical Results
Numerical results are shown for the case of an atom placed just at the middle point between two gold spheres with radii 40 [nm]. The atom is modeled as a two-level quantum system with parameters given in ref. [2]. For the dielectric function of gold a Drude model has been used [14].

![Relative Fluorescent Power](image)

Fig. 1. Fluorescent power normalized to the isolated atom as a function of the distance \( s \) between the atom and the spheres edges for three values of the laser intensity at the resonance frequency.
Figure 1 shows the fluorescent power (normalized to that of the isolated atom) as a function of the distance between the atom and the edges of the spheres. It shows that the power decreases as the atom moves away from the spheres. Due to the strong field enhancement, the fluorescent enhancement effect predominates over the quenching effect even for small distances. Furthermore, it can be seen that the greater the intensity of the incident laser the smaller is the normalized power, in agreement with equation (1).

In figure 2 we show the power as a function of the detuning factor $\delta$ for cases $\eta_o/\gamma_o = 0.5, 1$ and 2, where $\eta_o$ and $\gamma_o$ are the Rabi frequency and transition rate for the isolated atom. The local field enhancement due to spheres polarization increases the power broadening [11] and therefore this ratio increases nearly quadratically with detuning.

![Figure 2](image_url)

**Fig. 2.** Fluorescent power enhancement versus detuning for an atom placed at a distance of 0.04 nm from two gold spheres with radii 40 nm for $\eta_o/\gamma_o = 2$ (solid curve), 1 (dotted curve) and 0.5 (dashed curve)

**4. Conclusions**

By means of a semi classical model we have studied the influence of nanoparticles in the surroundings of atoms, over the atomic fluorescent radiated power, when the atom is under a laser with frequency close to resonance.

This model may be used to interpret results from experiments where atoms embedded in an optical environment formed by nanoparticles clusters are excited by an external field yielding combined effects of enhancement and fluorescence quenching.

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