Simulation of spontaneous emission in hyperbolic media

A R Gubaydullin¹²³, K A Ivanov², M A Kaliteevski¹²³

¹ St. Petersburg Academic University, Khlopina 8/3, 194021, St. Petersburg, Russia
² ITMO University, Kronverkskiy pr. 49, 197101, St. Petersburg, Russia
³ School of Engineering and Computing Sciences, Durham University, Durham DH13LE, UK

e-mail: gubaydullin.azat@gmail.com

Abstract. We study the probability of spontaneous emission enhancement of the dipole emitter imbedded in the stratified metal-dielectric metamaterial, characterized by the hyperbolic isofrequency surface. We analyze the performance of finite-difference time-domain (FDTD) method implementations for a 3D simulation of the spontaneous emission of the dipole source embedded in the metal dielectric stratified media. We discuss boundary conditions applicability.

1. Introduction

Metamaterials have attracted significant scientific interest because they offer a way to engineer many fascinating applications, including negative refractive index materials [1], metamaterial antennas [2], and enhancement of spontaneous emission [3,4]. Spontaneous emission is in particular interest since it plays an essential role in many phenomena of light–matter interaction. Experimentally the enhancement of spontaneous emission and superradiance are demonstrated in periodic dielectric nanostructures [5].

Commonly studied model of a hyperbolic metamaterial is a stratified structure which consists of infinite number of metal–dielectric periods. This system is expected [4] to demonstrate special optical properties emanating primarily from hyperbolic dispersion of isofrequency surface at certain frequency ranges. According to the conventional effective-medium approach a metal–dielectric nanostructure for certain frequency ranges can be analyzed as a uniaxial anisotropic medium, which effective permittivity tensor according to the conventional effective-medium approach a metal–dielectric nanostructure for certain frequency ranges can be analysed as a uniaxial anisotropic medium, which effective permittivity tensor $\varepsilon_{\text{eff}}$ of the diagonal form:

$$
\varepsilon_{\text{eff}} = \begin{pmatrix}
\varepsilon_{\perp} & 0 & 0 \\
0 & \varepsilon_{\perp} & 0 \\
0 & 0 & \varepsilon_{||}
\end{pmatrix},
$$

where components $\varepsilon_{\perp}$ and $\varepsilon_{||}$, depend on the thickness of layers $d_m$ (metal) and $d_d$ (dielectric) and corresponding permittivity, are given by:

$$
\varepsilon_{\perp} = \frac{d_m\varepsilon_m + d_d\varepsilon_d}{d_d + d_m}, \quad \varepsilon_{||} = \frac{(d_m + d_d)\varepsilon_d\varepsilon_m}{d_m\varepsilon_d + d_d\varepsilon_m}.
$$

And dispersion relation of a layered metal–dielectric nanostructure is given by:
For a certain frequency ranges and a certain ratio of the thickness of layers can demonstrate a hyperbolic shape of isofrequency surface.

Nowadays, numerical modeling is being widely used in research activities as well as in engineering. The finite-difference time-domain (FDTD) method was first introduced by Kane Yee in 1966 [6]. And nowadays, when highly-efficient computer facilities have been developed FDTD method attracted significant scientific interest for electromagnetic phenomena simulations. There are a number of applications based on the FDTD method applied to solve parabolic equations which provide a numerical solution of wave propagation in anisotropic, nonlinear, and dispersive media. To perform calculations of spontaneous emission we use the commercial software simulation package Lumerical FDTD Solutions.

2. Theory and model

The concept of the radiative decay enhancement in a cavity first proposed by Purcell in 1946 [7] within the framework of nuclear magnetic resonance and is known as the Purcell effect. This effect is described by the Purcell factor, which in the broad sense is defined by the ratio:

$$F_p = \frac{\tau_{\text{bulk}}}{\tau_{\text{cav}}}$$  

between lifetimes of spontaneous emission of a point light source in the infinite homogeneous medium to that inserted into a resonant cavity. According to the Fermi golden rule, the radiative decay rate $1/\tau$ in a homogeneous lossless medium is defined by:

$$\frac{1}{\tau} = \frac{2\pi}{\hbar} \sum_{k,\sigma} |\langle f | H_{\text{int}}(k, \sigma) | i \rangle|^2 \delta(h \omega_{k,\sigma} - h\omega)$$  

where $\langle f | H_{\text{int}}(k, \sigma) | i \rangle$ is the matrix element between the initial state $| i \rangle$ and the final state $| f \rangle$ $h\omega$ is the transition energy with the emission of the photon $(k, \sigma)$, characterized by the wave vector $k$ and polarization $\sigma$. The emitter is commonly assumed as an atom and is initially assumed to be in an excited state, and can be characterized by the dipole moment $d$, henceforth simply referred to as either dipole or emitter. For the quantum transition characterized by dipole moment $d$ the Fermi golden rule can be written:

$$W = \frac{2\pi}{\hbar} |\langle f | Ed | i \rangle|^2 \rho$$  

Where in the matrix element $\langle f | Ed | i \rangle$ for the transition between states $| i \rangle$ and $| f \rangle$, $E$ describe spatial distribution of the electric field of the mode, and $\rho$ the density of states. The density of photonic states in hyperbolic medium diverges providing an enhancement of the spontaneous emission [4], which allows to tune the emission properties of the light source, potentially useful for a variety of applications. In essence, the problem is leading to calculation of the distribution of the electric field of the mode in the structure and the density of states.

The FDTD method solves Maxwell's equations on a discrete spatial and temporal grid applying specific boundary conditions which should be accurately chosen depending on the particular model. The FDTD solver supports a range of boundary conditions (BC), such as perfectly matched layer (PML) [8,9], periodic [10], and Bloch BC (useful for oblique incidence in the periodic systems). PML BC absorb electromagnetic waves incident upon them. Since we assume our structure infinite in $x$ and $y$ directions we perform FDTD calculations using PML BC on these boundaries.

Periodic BC's are useful when studying periodic systems, they afford calculating the response of the whole system by only simulating one unit cell. Periodic BC's copy the electromagnetic fields which occur at one boundary of the simulation area and inject them at the opposite boundary. Periodic BC's provide fast simulation, however, their applicability is limited only for completely periodic systems, when both the physical structure and the electromagnetic fields are periodic.

In this paper we consider a periodic configuration of alternating silver and dielectric layers with permittivity $\varepsilon_{m,d}$ and layer thicknesses $d_{m,d}$ respectively, with a dipole emitter imbedded in the central dielectric layer. The system is shown schematically on figure 1. The permittivity of silver is defined by

$$\frac{k_x^2 + k_y^2}{\varepsilon_{m}} + \frac{k_z^2}{\varepsilon_{p}} = \left(\frac{\omega}{c}\right)^2$$  

(3)
the Drude model: \( \varepsilon_m = \varepsilon_\infty - \frac{\omega_p^2}{\omega(\omega+i\gamma)} \) which was fitted to a particular frequency range of the Johnson and Christy data [11] with the following parameters: \( \varepsilon_\infty \approx 4.96 \), the plasma frequency \( \omega_p \approx 8.98 \text{ eV} \), and \( \gamma \approx 0.018 \text{ eV} \). In complex metamaterials, the effective plasma frequency can be significantly lower than that which belongs to bulk metal [12]. The dielectric constant \( \varepsilon_d = 1 \), the structure period \( D = d_m + d_d = 30 \text{ nm} \), and the ratio of the thickness of silver and dielectric layers is varied \( \eta = \frac{d_m}{d_d} \). It is convenient to study two dipole orientations parallel and perpendicular to layers of the structure.

![Figure 1. The dipole emitter centered in the stratified metal-dielectric structure.](image)

3. Results and discussion

The Purcell factor or the decay rate enhancement is commonly defined as a ratio between the power emitted by a dipole source imbedded into a photonic structure to the power emitted by the dipole in a homogeneous environment. To calculate the Purcell factor Lumerical proposed several options. The first is an estimation of the Purcell factor by using the “dipolepower” command, which returns the power actually radiated in the environment.

We study hyperbolic media as a stratified structure which consists of infinite number of metal-dielectric periods. Therefore we start our modelling with periodic BC’s in the \( z \) direction, results calculated by using the dipolepower command are illustrated on figure 2 (a,b). An enhancement of the spontaneous emission and a sharp dip near the Surface Plasmon Polariton (SPP) [13,14] frequency could be seen, as it was presented in [4]. Interesting to note that our results are in good agreement with results presented in [4], although authors used approach proposed by F.de Martini [15]. The original approach [15] is based on the integration over the incidence angle inside the light cone, a direct application of this approach to our metal-dielectric system was illustrated in [16].

![Figure 2. The Purcell Factor of the stratified structure calculated with “dipolepower” command in the FDTD Solutions package of Lumerical. The period of the structure is fixed \( D = 30 \text{ nm} \), while the ratio of the thickness of layers \( \eta = \frac{d_m}{d_d} \) is varied. Two orientations of the dipole emitter: (a) perpendicular and (b) parallel with respect to the interface.](image)
We should note that periodic BC's are applicable only for infinite models, in the case of finite structures (as in experiment) application of periodic BC's may lead to inaccuracies in the analysis of systems of finite size, as was demonstrated in [17]. Moreover, since we are interested in simulating a system with one single dipole source the system can not be simulated as periodic, since it does not have one dipole per each period, therefore hereinafter we will focus on the structure which is constructed from 6 periods with a dipole source embedded in the central dielectric layer, which results obtained with “dipolepower” command are presented on figure 3.

The second method is based on the Green's function formulation [18,19] which relies on the fact that the emission rate is proportional to the local density of optical states. Lumerical has implemented Green's function approach to calculate the spontaneous decay rate for a two-level quantum system at $r_0$ and $\omega$ by:

$$\gamma = \frac{2\omega}{3\hbar\varepsilon_0}|\mu|^2\rho_x(r_0, \omega),$$

where the partial local density of states along Z axes can be obtained from the imaginary part of Green's function:

$$\rho_x(r_0, \omega) = \frac{6\omega}{\pi c^2}\text{Im}\{G_{xx}(r_0, r_0; \omega)\}$$

Results of FDTD simulation obtained by using the Green's function technique, implemented in the FDTD Solutions package of Lumerical, are illustrated on figure 3 by dashed green curve. The frequency dependence of the Purcell factor calculated by the “dipolepower” method is illustrated on figure 3 by red solid curve.

![Figure 3. The frequency dependence of the Purcell factor of the stratified structure calculated by the “dipolepower” method is illustrated by the red solid curve. The Purcell Factor calculated by using the Green's function technique - dashed green curve, and by using the “box of monitors” technique - black solid curve. The period of the structure is fixed D=30 nm, the ratio of the thickness of layers $\eta = 1$. The dipole orientation is assumed to be parallel to layers.](image-url)

However, the dipolepower technique and the Green's function formulation only work if the dipole is in a lossless dielectric medium. If the dipole is imbedded in a lossy media it is recommended by Lumerical to measure the power flow out of a small box of monitors surrounding the source in the environment. We also perform calculation using a transmission box of monitors enclosing our model. The dependence of the Purcell factor on frequency obtained by using the box of monitors is illustrated by black solid curve on figure 3.

The comparison of results obtained by all three methods are illustrated on figure 3. As one can note the method based on the dipolepower function and the method based on the Green's function illustrate similar estimations of the Purcell factor, however, as was noted by Lumerical those two methods are not applicable for a lossy media. The method based on measurement of the power flow through a small box of monitors illustrates values of the Purcell factor, which for certain frequencies are by several orders of magnitude smaller than results predicted by the Green's function method. We should
note that results depend on many simulation parameters and require a lot of computational effort for convergence tests, and variation of simulation parameters leads to strong changes of calculated results, that confirms the need to develop a stable approach for calculating the probability of spontaneous emission in arbitrary layered structures.

In our recent work we have developed a rigorous self-consistent procedure of quantization of electromagnetic field for inhomogeneous media (S-quantization) [17], which provide accurate calculation of the probability of spontaneous emission for arbitrary, asymmetric metal/dielectric structures [20]. S-quantization also allows analysis of the Purcell effect in the waveguide mode regime [21].

4. Conclusions
We performed simulation of the hyperbolic metamaterial constructed from one-dimensional metal-dielectric nanostructure. We study the probability of spontaneous emission enhancement of the dipole emitter imbedded in the stratified metal-dielectric metamaterial by FDTD simulation methods. We have shown that the probability of spontaneous emission depends on the simulation parameters and applied formalism. Therefore it is of particular interest to develop reliable approach to calculate the Purcell factor for arbitrary structures.

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