Efficient optical modeling of spontaneous emission in a cylindrically layered nanostructure

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Abstract: We present an efficient optical model to study spontaneous emission in a cylindrically layered nanostructure. The total emission power of an emitter in the nanostructure is efficiently calculated. A formula is derived to calculate the lateral-surface emission power. As examples of practical interest, spontaneous emission properties, including radiative transition rate of the emitter, the assignment of the emission to lateral-surface emission and waveguided emission, are comprehensively studied at the first time for an isolated ZnO nanowire and a ZnO/SiO2 nanocable.

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References and links

1. Introduction

One-dimensional semiconductor nanostructures, such as nanowires (NWs) [1-2], core/shell nanocables (NCs) [3-5] and nanotubes (NTs) [6], have been under intensive research in recent years owing to their impressive functionalities in building up novel nanoscale photonic elements such as light-emitting diodes (LEDs), lasers and biosensors. These nanostructures can be approximately considered as cylindrically layered structures and their optical properties depend on the structural parameters. For example, the quantum efficiency of a NW LED should be different from that of a planar multilayered thin film LED since radiative transition rates of the emitter in these structures are distinct [7-9]. In addition, the assignment of the total emission power to the lateral-surface emission and the waveguided emission (propagating along longitudinal axis of the nanostructure) also depends on the size of the nanostructure. These properties are critical to NWs or NCs for LED and fluorescence labeling applications. In this paper these issues will be systematically studied.

The theoretical description of spontaneous emission (SE) in a cylindrically layered nanostructure is based on the classical electromagnetics with the emitter modeled as an incoherent electric dipole (point source) running at a constant current [8]. The radiative transition rate of the quantum emitter and its lateral-surface emission efficiency can be characterized through the total emission power and lateral-surface emission power radiated by the electric dipole [8,9]. Spontaneous emission and the response of a classical dipole source in cylindrical structures have been studied for decades from various angles by using different methods [10-16], including cylindrical wave decomposition method [10,13,14], a combination of Fourier integral and multipole methods [15] and the three-dimensional finite difference time domain (3D-FDTD) method [16]. However, the methods in refs.[10,13-15] need special treatments for the waveguide modes in the cylindrical structure and are inefficient for the evaluation of the total emission power. The 3D-FDTD method is also inefficient. In this paper, we use a simple technique without any special treatment for the waveguide modes to calculate the total emission power and give an explicit formula to calculate the lateral-surface emission power. The theory will be given in Section 2. SE properties, including the radiative transition rate and the assignment of the emission to lateral-surface emission, are studied in Section 3 for an isolated ZnO NW and a ZnO/SiO2 NC. Conclusion is addressed in Section 4.

2. Theory

The geometry of a cylindrically multilayered nanostructure is shown in Fig. 1. An emitting layer is sandwiched between two stacks of shells, i.e., $N$ outer shells and $M$ inner shells. The emitting medium and the outer most medium (e.g. air) are assumed to be non-absorbing at the emitting wavelength while the other shells can be either transparent or absorptive. The quantum emitter is modeled as an incoherent classical electric dipole (point source) with a constant current. The orientation of the electric dipole is parallel to the electric field of the excitation in photoluminance (PL) [17, 18] and is random in space in electrolumiance. The SE properties of the emitter can be characterized by the total emission power $F$, the lateral-surface emission power $U$ and waveguided emission power $W$ (all are normalized by the total emission power of the dipole in infinite medium) of the dipole in the cylindrically layered nanostructure. If the materials are all lossless, one has $F = U + W$. As a consequence of Fermi’s golden rule, the radiative transition rate is optical-environment dependent as [8,9]

$$\Gamma_r = F \cdot \Gamma_r^0$$

(1)

where $\Gamma_r^0$ and $\Gamma_r$ are the radiative transition rate in the infinite medium and the cylindrically layered media, respectively. Assuming that the nonradiative transition rate in the cylindrically layered media is $\Gamma_{nr}$, the internal quantum efficiency $\eta_i$, lateral-surface emission efficiency $\eta_U$.
and waveguided emission efficiency \( \eta_w \) are obtained as
\[
\eta_F = \Gamma_z, G, \Gamma_\omega \rightleftharpoons F \eta_0 / \left[ F \eta_0 + (1 - \eta_0) \right] \tag{2}
\]
\[
\eta_U = \eta_F \cdot U / F = U \eta_0 / \left[ F \eta_0 + (1 - \eta_0) \right] \tag{3}
\]
\[
\eta_w = \eta_F \cdot W / F = W \eta_0 / \left[ F \eta_0 + (1 - \eta_0) \right] \tag{4}
\]
where \( \eta_0 = \Gamma_z \) is the initial internal quantum efficiency. In the cylindrically layered media, the electric field induced by an internal dipole can be written as
\[
E = E^i + E^r \tag{5}
\]
where \( E^i \) and \( E^r \) are the electric field of the dipole in the infinite medium and the reflected electric field by the other shells, respectively. The total emission power can be obtained as [8]
\[
F^u = 1 - \alpha \cdot \text{Im} \left( E^i \cdot \left( E^r, r' \right) \right)/(\omega \mu_0 \epsilon_0 / \omega \pi) \tag{6}
\]
where \( \mu_0, \epsilon_0, k, \Theta, \tilde{\alpha}, \) and \( \tilde{r}' \) are the permeability, angular frequency, wavenumber in the emitting medium, dipole moment, orientation and location of the dipole, respectively. Here \( \text{Im} \) stands for the imaginary part of \( () \) and the superscript of \( F \) denotes the orientation. To calculate \( F \) and \( U \), we decompose the z-component of \( E^r \) and \( H^r \) (magnetic field of the dipole in the infinite medium) in terms of cylindrical waves in polar coordinates \( (\rho, \theta, z) \) [10,19]
\[
\begin{bmatrix}
E_{z}^0 \\
H_{\rho}^0
\end{bmatrix} = \sum_{n=0}^{\infty} e^{n\theta} \int dk, \tilde{a}_{0n}, H, (k, \rho) e^{i\tilde{a}, z} \quad (\rho > \rho^*)
\]
\[
\begin{bmatrix}
E_{z}^0 \\
H_{\rho}^0
\end{bmatrix} = \sum_{n=0}^{\infty} e^{n\theta} \int dk, \tilde{b}_{0n}, J, (k, \rho) e^{i\tilde{a}, z} \quad (\rho < \rho^*)
\]
where
\[
\begin{align}
\tilde{a}_{0n} &= -\frac{\Theta}{8\pi\epsilon_0 \omega \left( \tilde{z} k^2 + \frac{\partial}{\partial \tilde{z}} \right)} \left( \tilde{z} k^2 + \frac{\partial}{\partial \tilde{z}} \right) \tilde{\alpha} e^{-i\theta - \frac{\tilde{a}}{\tilde{z}}} J, (k, \rho^*) \tag{8.a}
\end{align}
\]
\[
\begin{align}
\tilde{b}_{0n} &= -\frac{\Theta}{8\pi\epsilon_0 \omega \left( \tilde{z} k^2 + \frac{\partial}{\partial \tilde{z}} \right)} \left( \tilde{z} k^2 + \frac{\partial}{\partial \tilde{z}} \right) \tilde{\alpha} e^{-i\theta - \frac{\tilde{a}}{\tilde{z}}} J, (k, \rho^*) \tag{8.b}
\end{align}
\]
Here \( \epsilon, k_\rho = \sqrt{k^2 - k_z^2} \) and \( k_z \) are the permittivity, the radical and \( z \) components of the wavenumber, respectively. \( J, \) and \( H, \) denote the Bessel function and Hankel function of first kind at order \( v \). The two stacks of shells are considered as two “black” shells characterized by the total downward reflection matrix \( N_{0v}, \) the total upward reflection matrix \( M_{0v}, \) and the total transmission matrix \( T_{Nv}. \) Then the z component of the reflected field in the emitting layer and the transmitted field in the outermost medium can be obtained as
\[
\begin{bmatrix}
E_{z} \\
H_{\rho}
\end{bmatrix} = \sum_{n=0}^{\infty} e^{n\theta} \int dk, \tilde{a}_{0n}, H, (k, \rho) + \tilde{b}_{0n}, J, (k, \rho) e^{i\tilde{a}, z} \tag{9}
\]
\[
\begin{bmatrix}
E_{z} \\
H_{\rho}
\end{bmatrix} = \sum_{n=0}^{\infty} e^{n\theta} \int dk, \tilde{a}_{0n}, H, (k, \rho) e^{i\tilde{a}, z} \tag{10}
\]
remarkable features can be observed in Fig. 3(a). First of all, located at origin of the NW and with orientation directions are denoted as normalized total emission powers of the emitter orienting along the radical, azimuthal and z direction. Several remarkable features can be observed in Fig. 3(a). First of all, $F^r$ is smaller than $F^z$ by nearly an order of magnitude for the ZnO NW with small radius. As the radius of the NW further

\[
\bar{a}_{0v} = [\bar{I} - M_{0v}, N_{0v}] [M_{0v}, \bar{b}_{00v} + \bar{a}_{00v}] - \bar{a}_{00v} 
\]

\[
\bar{b}_{0v} = [\bar{I} - N_{0v}, M_{0v}] [N_{0v}, \bar{a}_{00v} + \bar{b}_{00v}] - \bar{b}_{00v} 
\]

\[
\bar{a}_{Nv} = T_{Nv}(\bar{a}_{0v} + \bar{b}_{00v}) 
\]

The field transverse to the z-direction can be readily obtained from the z component of the field. For the detailed derivations of $N_{0v}, M_{0v}$ and $T_{Nv}$, we refer the readers to ref. [10].

2.1 Calculation of the total emission power $F$

As seen from Eqs. (6) and (9), the calculation of $F$ involves the evaluation of an integral which has pole singularities, physically corresponding to the guided modes in the cylindrically layered structure. This is similar to the case of a planar multilayered structure [20] and a direct evaluation of the integral in Eq. (9) tends to fail. These poles are located in the real axis of $k_x$ or in the first quadrant of the complex plane of $k_z$. Here, we extend the integration domain in Eq. (9) to the complex plane of $k_z$ and use the Cauchy integral theorem to perform the integration. We select an integration path in the fourth quadrant such that the domain enclosed by the new path and the real axis of $k_z$ does not contain any pole. The new path shown as the dashed line in Fig. 2 consists of a semicircle in the fourth quadrant of the complex plane and a straight line along the real axis. The integral in Eq. (9) can be written as

\[
\left[ \begin{array}{c} E^z \\ H^z \end{array} \right] = 2 \sum_{v=0}^{\infty} e^{i\theta_{v0}} \left( \int_{C_R} + \int_{-\infty}^{\infty} \right) \bar{a}_{0v} H^{(1)}_{v} (k_x, \rho) + \bar{b}_{0v} J_{v} (k_x, \rho) e^{i k_z z} dk_z 
\]

where $k_x$ is the radius of the semicircle $C_R$. $k_x$ should be large enough so that the semicircle can bypass all the poles of the integrand. In this way, the integration is efficiently evaluated.

2.2 Calculation of the lateral-surface radiation power

The normalized lateral-surface emission power $U$ is the power leaving the cylindrically layered structure from the lateral surface. Thus it can be expressed as

\[
U = \int_{0}^{\frac{\pi}{2}} \rho d \theta \int_{-\infty}^{\infty} \frac{1}{2} \text{Re} \left( E_{\rho} \times H_{\rho} \right) / L_0 
\]

where $L_0 = \sqrt{\mu / \epsilon_{out} k_{out}^2} / 4\pi$ is the normalization constant and $\text{Re} ()$ stands for the real part of $()$. Here $\epsilon_{out}$ and $k_{out}$ are the permittivity and wavenumber of the outermost medium, respectively. After some algebra manipulation, Eq. (13) can be written as

\[
U = \frac{\alpha P}{L_0} \sum_{v=0}^{\infty} \int_{0}^{\frac{\pi}{2}} \int_{-\infty}^{\infty} \text{Re} \left[ \epsilon_{out} E^{(1)}_{\rho}(k_x, \rho) H^{(1)}_{\rho}(k_x, \rho) a_{\rho v}(1) - i \mu H^{(1)}_{\rho}(k_x, \rho) H^{(1)}_{\rho}(k_x, \rho) a_{\rho v}(2) \right] / k_x 
\]

If the outermost medium is lossless, $U$ will not depend on $\rho$.

3. Results and Discussion

In this section, the radiative transition rate, the lateral-surface emission efficiency $\eta_U$ and the waveguided emission efficiency $\eta_W$ are studied for an isolated ZnO NW and a ZnO/SiO$_2$ NC at a wavelength of 556 nm, i.e. the PL peak of a broadband ZnO NW LED [2]. The cross-section of the ZnO NW, which is actually hexagonal, is treated approximately as circular here. In our simulation, an initial internal quantum efficiency $\eta_i=0.85$ is used according to ref. [21]. The refractive indices of 2.45 and 1.46 are used for ZnO and silica, respectively. Below, the normalized total emission powers of the emitter orienting along the radical, azimuthal and z directions are denoted as $F^r$, $F^\theta$ and $F^z$, respectively. Similar definitions also apply for $U$.

3.1 SE of an isolated ZnO NW

Figure 3(a) shows the dependences of $F^r$ and $U^r$ on the radius of the ZnO NW for the emitter located at origin of the NW and with orientation $x$ (radical, azimuthal or z direction). Several remarkable features can be observed in Fig. 3(a). First of all, $F^r$ is smaller than $F^z$ by nearly an order of magnitude for the ZnO NW with small radius. As the radius of the NW further
decreases, $F'$ has a limiting value of 0.033. This limiting value depends on the refractive index of the NW and becomes smaller as the refractive index increases as shown in the inset of Fig. 3(a). Since the radiative transition rate is proportional to $F$ (c.f. Eq. (1)) and radiative transition and non-radiative transition are two competing processes, the internal quantum efficiency of the emitter orienting along the radial direction (below we will call it as $\rho$-oriented emitter) is significantly low (c.f. Eq.(2)). Thus for the NWs with small radii the $z$-oriented emitters are much more efficient. Secondly, there are two critical radii of the NW, i.e. 45 nm and 95 nm for the $\rho$-oriented emitter and $z$-oriented emitter, respectively. For the radius less than 45nm, $\eta_w$ for both kinds of emitter is less than 1%. But for a radius between 45nm and 95nm, only $\eta_w$ for $z$-oriented emitter is less than 1%. As the radius exceeds 45 nm, $\eta_w$ of the $\rho$-oriented emitter first increases and then oscillates. Here a larger critical radius for the $z$-oriented emitter is due to the fact that the emission by the emitter located at the origin of the NW can not couple to the fundamental $HE_{11}$ mode but have a good coupling to waveguide mode starting from TM$_{01}$ mode which is cutoff for the radius smaller than 95 nm. Once the radius exceeds 95 nm, an abrupt change of $U$ occurs for the $z$-oriented emitter.

Fig. 3(b) shows the variation of $\eta_U$ and $\eta_w$ as the change of the radius of the ZnO NW for the emitter located at the origin of the NW. As observed in Fig. 3(b), $\eta_U$ of the $z$-oriented emitter is nearly four times larger than that of the $\rho$-oriented emitter for the NWs with the radii smaller than 30 nm. For the NW with a radius of 95 nm, $\eta_w$ of the $\rho$-oriented emitter reaches the maximum value of 0.73, approximately 5.5 times larger than $\eta_U$. We have also studied the SE properties of the emitter located near the boundary of the NW. Figure 3(c) shows the variation of $\eta_U$ and $\eta_w$ as the change of the radius for the emitters located near the boundary of the NW. Different from Fig. 3(b), $\eta_w$ of the emitters with different orientations have the same critical radius of 45 nm, below which $\eta_w$ is less than 1%. This is because the emission for the emitter near the boundary of the NW with any orientation (including along $z$ direction) couples to the waveguided emission starting from the fundamental $HE_{11}$ mode.

From Fig. 3(a)-(c), one sees that for the ZnO NW with a radius less than 45 nm almost no waveguided emission can be observed. For the NWs with such small radii, $\eta_U$ of the $z$-oriented emitter is much larger than that of the $\rho$-oriented or $\theta$-oriented emitter. Since in PL the orientation of the emitter is parallel to the electric field of the excitation [14, 15], $\eta_U$ by PL will be sensitive to the polarization of the excitation light.

3.2 SE of a ZnO/SiO$_2$ nanocable

Semiconductor NWs with coaxial shells of different kinds of materials or dopants are interesting owing to their potential applications [3-5]. Semiconductor NWs with silica shell can reduce non-radiative recombinations and protect them from aggregation, leading to an improved chemical stability [4]. In this subsection, the SE properties of a ZnO/SiO$_2$ nanocable (NC) will be studied. As seen in Fig. 3(a) and 3(b), for the emitter located at origin of the isolated ZnO NW with a radius between 45 nm and 95 nm, $\eta_w$ of the $\rho$-oriented emitter exists while that of the $z$-oriented emitter is almost zero. Thus we expect that for a ZnO/SiO$_2$ NC with a small core radius and a suitable shell, $\eta_w$ of the $\rho$-oriented or $\theta$-oriented emitter will be much larger than that of $z$-oriented emitter, meanwhile $\eta_U$ of $z$-oriented emitter is larger. Thus for such a coaxial ZnO/SiO$_2$ NC, $\eta_w$ and $\eta_U$ will show opposite polarization anisotropy in PL. To validate this, we study the SE properties of the ZnO/SiO$_2$ NCs with a core radius of 10 nm and silica shell of various thicknesses. Fig. 3(d) shows the variation of $\eta_U$ and $\eta_w$ as the change of the shell thickness for the emitters located near the boundary of the ZnO core with different orientations. Similar results are obtained for the emitter located at the origin of ZnO core. The curves for the $\rho$-oriented emitter and $\theta$-oriented emitter are almost identical due to the small radius of the ZnO core. Two critical shell thicknesses, 50 nm and 180 nm, are observed for the $\rho$-oriented emitter and $z$-oriented emitter, respectively. Below the critical thickness, $\eta_w$ of the emitter with any orientation is less than 1%. While for the shell thickness between 50 nm and 180 nm, the waveguided emission exists for $\rho$-oriented or $\theta$-oriented emitter but is nearly zero for the $z$-oriented emitter. In the meantime, $\eta_U$ of the $z$-oriented emitter is much larger than that of the $\rho$-oriented emitter.
For the ZnO NW or ZnO/SiO₂ NC with a finite length, an optical cavity is formed in the NW due to the facet reflection. In this case, the facet emission power roughly equals the waveguided emission power due to the following two reasons (i) the cavity effect is weak since the power reflection coefficient is less than 0.177 (as roughly given by \((n_{ZnO} - n_{air})^2/(n_{ZnO} + n_{air})^2)\) (ii) the waveguided emission for the infinitely long NW equals the facet emission of the emitter located in the middle of the node and antinode of the standing wave formed in the cavity. The facet emission of the NW is the averaged emission by the emitters located within the node and antinode of the standing wave.

4. Conclusions

In conclusion, we have presented an efficient and accurate optical modeling of SE for cylindrically layered nanostructures. The total emission power is efficiently calculated by selecting a new integration path in the complex plane. An explicit formula has been derived to calculate the lateral-surface emission power. The SE properties of an isolated ZnO NW and a ZnO/SiO₂ NC are comprehensively studied. The radiative transition rate of the emitter in the NW depends strongly on its orientation. The \(z\)-oriented emitters are usually much more efficient than the \(\rho\)-oriented or \(\theta\)-oriented emitters for a NW with a small radius since the transition rate of the latter is smaller than that of the former by nearly one order of magnitude. There is a critical value of the radius of the NW below which the waveguided emission efficiency is almost zero. For a ZnO/SiO₂ nanocable with a suitable silica shell thickness the lateral-surface emission and waveguided emission show opposite polarization anisotropy.

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