Radiation of a Point Dipole in "Spherical Semiconductor Quantum Dot + Spherical Metal Nanoparticle" Structure (theory)

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***Abstract* — A model is proposed allowing considering radiation of semiconductor quantum dot (QD) in a vicinity of metal nanoparticle (NP) as radiation of an ensemble of unit cells, from which the QD consists. It is shown that each unit cell of QD can be considered as a separate radiating point dipole. Relations are obtained between the coefficients of multipole expansions of electromagnetic fields in two spherical coordinate systems centered in the semiconductor QD and the metal NP. The relations allow using separately spherical symmetry of the QD and the NP despite the "QD + NP" structure as a whole is not spherically symmetric. It is shown, that contribution of a particular QD unit cell into the total QD radiation is proportional to the value of the envelope exciton wave function in corresponding QD node. Summing contributions from all QD unit cells makes possible to find characteristic radiative and nonradiative times in "QD + NP" structure and evaluate their decrease relatively to that of isolated QD due to resonant excitation of dipole and higher order multipole plasma oscillations in metal NP. For the metal NP not only contribution of free carriers to its dielectric function is taken into account (Drude term) but that of bound carriers too due to possible interband transitions in real metals in the actual frequency range. Besides, to cover the cases of extremely small metal NPs (less then 10 nm in size) a spatial dispersion in NP dielectric function is also accounted**.

***Index Terms* — metal nanoparticle, multipole amplitudes, point dipole, radiation scattering, semiconductor quantum dot**

I. INTRODUCTION

For the wide practical perspectives the intense theoretical studies of radiation characteristics of point emitters (dipoles) placed near small metal particles had been carried out in last decades. It had been shown that due to resonant excitation of plasma oscillations in such particles the characteristic times of both radiative and nonradiative losses of the emitters depend on the distance between the emitter and the metal particle, as well as on particle size. As a result, the efficiency of the emitter radiation depends non-monotonically on the distance [1-3]. Considering the emitter as a point dipole is completely justified in the case of isolated atom or molecule luminophore. However, by studying radiation of semiconductor QD in a vicinity of metal NP the QD *a priori* can not be considered as a point dipole (especially at comparable QD and NP sizes). Therefore, all previous theoretical approaches in fact can not be directly applied to this system. The goal of this work is just to develop a model allowing calculations for the case of semiconductor QDs in a vicinity of metal NPs.

We have shown that despite semiconductor QD as a whole can not be considered as a point dipole, its emission actually can be described as emission of a set of point dipoles with unit cell size as "point" size. Therefore, to calculate the emission characteristics of semiconductor QD in a vicinity of the metal NP it is necessary to consider previously the contribution to the emission of arbitrary unit cell in QD, and then integrate the contributions from all QD unit cells with corresponding weight factors in the form of the product of the electron and hole size-quantized envelope wave functions. In this paper, we have considered the first part of this problem, which is reduced to the calculation of the electromagnetic fields of a point dipole radiating inside the semiconductor QD in the case when QD is located in an immediate vicinity of metal NP. The consideration is made in a single scattering approximation. Multiple scatterings of electromagnetic field in "QD + NP" structure could be accounted too using the obtained relations between coefficients of the field multipole expansions in two different spherical coordinate systems with the centers in QD and NP.

II. GROUNDS FOR THE MODEL OF QD RADIATION AS RADIATION OF AN ENSEMBLE OF OSCILLATING POINT DIPOLES INSIDE QD

Consider QD multi-electron system when one of the electrons is excited from a state associated with the valence band of bulk material, from which QD is formed, into the state associated with conduction band. In the

linear approximation, the solution of time-dependent Schrцdinger equation can be written in this case as follows:

$$\Psi(t) = \Psi_0 \exp(-\mathrm{i}\omega_0 t) + \Psi_1 \exp\left(-\mathrm{i}\,(\omega_0 + \omega)t\right), \quad (1)$$

where Ψ_0 is the wave-function of the ground state of multi-electron system with Hartree-Fock energy $\hbar\omega_0$, Ψ_1 is the correction to the wave function Ψ_0 due to specified perturbation, $\hbar\omega$ is the energy of the perturbation. Wave function Ψ_0 can be written as antisymmetrized product of one-electron orthonormal Wannier lattice-site functions of the valence band [4]:

$$\Psi_0(\tilde{\xi}_1,..\,\tilde{\xi}_{NK_{\alpha\sigma}}) = 1/\sqrt{(NK_{\alpha\sigma})!}\sum_p (-1)^{A_p}\hat{p}\prod_{\tilde{\mathbf{i}},\alpha,\sigma} b^V_{\tilde{\mathbf{i}},\alpha,\sigma}(..)\left\{\tilde{\xi}_1,..\,,\tilde{\xi}_{NK_{\alpha\sigma}}\right\}, \quad (2)$$

where $\hat{p}$ is the operator of a particular permutation from total $(NK_{\alpha\sigma})!$ permutations, at which $NK_{\alpha\sigma}$ space-spin electron variables $\left\{\tilde{\xi}_1, \tilde{\xi}_2, \ \ldots\ , \tilde{\xi}_{NK_{\alpha\sigma}}\right\}$ are distributed over $NK_{\alpha\sigma}$-fold product of valence-band lattice-site Wannier functions $b^V_{\tilde{\mathbf{n}},\alpha,\sigma}(\tilde{\boldsymbol{\xi}}) = b^V_{\tilde{\mathbf{n}},\alpha}(\tilde{\mathbf{r}})\chi_\sigma(s)$ as their arguments ($\tilde{\mathbf{r}}$ is the spatial and *s* the spin variable), A_p is multiplicity of the permutation *p*, *N* is the total number of lattice sites (nodes, unit cells) in the QD, $\tilde{\mathbf{I}}$ is one of these lattice sites ($\tilde{\mathbf{I}}$ in (2) runs over all *N* sites of QD crystal lattice), σ is a symbol of the electron spin state for a given degenerate state of the valence band, α is a symbol characterizing the degeneracy of the valence-band lattice-site Wannier functions over crystallographic directions (in cubic crystals all three crystallographic directions are degenerated), $K_{\alpha\sigma}$ is the multiplicity of degeneracy over electron spin and the crystallographic directions. Here and below, the tilde over the variables means that they are written in the coordinate system with the center in the semiconductor QD.

The wave function Ψ_1 can be written as a superposition of antisymmetrized $\chi^{\sigma\sigma'\alpha}_{\tilde{\mathbf{s}}\tilde{\mathbf{t}}}$-states in which one of the electrons of the multi-electron system is transferred from the localized in the site $\tilde{\mathbf{s}}$ $\alpha\sigma$-Wannier state of the valence band $b^V_{\tilde{\mathbf{s}},\alpha,\sigma}$ into the localized in the site $\tilde{\mathbf{t}}$ σ'-Wannier state of the conduction band $b^C_{\tilde{\mathbf{t}},\sigma'}$:

$$\Psi_1^{\sigma\sigma'}(\tilde{\xi}_1,\ldots,\tilde{\xi}_{NK_{\alpha\sigma}}) = \sum_{\tilde{\mathbf{s}},\tilde{\mathbf{t}},\alpha} C^{\sigma\sigma'\alpha}_{\tilde{\mathbf{s}}\tilde{\mathbf{t}}}\chi^{\sigma\sigma'\alpha}_{\tilde{\mathbf{s}}\tilde{\mathbf{t}}}(\tilde{\xi}_1,\ldots,\tilde{\xi}_{NK_{\alpha\sigma}}) \ . \quad (3)$$

The $C^{\sigma\sigma'\alpha}_{\tilde{\mathbf{s}}\tilde{\mathbf{t}}}$ coefficients are the probability amplitudes of such $\chi^{\sigma\sigma'\alpha}_{\tilde{\mathbf{s}}\tilde{\mathbf{t}}}$-states,

$$\chi^{\sigma\sigma'\alpha}_{\tilde{\mathbf{s}}\tilde{\mathbf{t}}} = 1/\sqrt{(NK_{\alpha\sigma})!}\sum_p (-1)^{A_p}\hat{p}\, b^C_{\tilde{\mathbf{t}},\sigma'}(..)\prod_{\tilde{\mathbf{i}},\gamma,\eta\neq\tilde{\mathbf{s}},\alpha,\sigma} b^V_{\tilde{\mathbf{i}},\gamma,\eta}(..)\left\{\tilde{\xi}_1,..,\tilde{\xi}_{NK_{\alpha\sigma}}\right\}. \quad (4)$$

In an analogy with [5,6] let us introduce the mean dipole moment $\mathbf{P}(\tilde{\mathbf{L}},t)$ of the lattice site $\tilde{\mathbf{L}}$, which arises due to above specified distortion of the QD multi-electron system,

$$\mathbf{P}(\tilde{\mathbf{L}},t) = \int \Psi^*(t)\,\hat{p}_0(\tilde{\mathbf{L}})\,\Psi(t)\,d\tilde{\Omega}_e \ , \quad (5)$$

where the integration is taken in all $NK_{\alpha\sigma}$ variables of multi-electron system, $\hat{p}_0(\tilde{\mathbf{L}})$ is the dipole moment operator for the site $\tilde{\mathbf{L}}$,

$$\hat{p}_0(\tilde{\mathbf{L}}) = e\sum_i (\tilde{\mathbf{r}}_i - \tilde{\mathbf{L}})T(\tilde{\mathbf{r}}_i - \tilde{\mathbf{L}}) \ . \quad (6)$$

In this formula the sum is taken over all *N* spatial variables of the electron subsystem, function $T(\tilde{\mathbf{r}}_i - \tilde{\mathbf{L}})$ equals zero if spatial variable $\tilde{\mathbf{r}}_i$ turns out to be outside the unit cell at the node $\tilde{\mathbf{L}}$ and $T(\tilde{\mathbf{r}}_i - \tilde{\mathbf{L}}) = 1$ if $\tilde{\mathbf{r}}_i$ is within the unit cell.

Carrying out integration in (5) for a chosen particular case of spin states σ and σ' (with an account for the electron-hole exchange interaction the perturbation energy $\hbar\omega$ can depend on these spin states), the following expression could be obtained:

$$\mathbf{P}(\tilde{\mathbf{L}},t) = e\sum_{\tilde{\mathbf{s}},\tilde{\mathbf{t}},\alpha} C^{\alpha}_{\tilde{\mathbf{s}}\tilde{\mathbf{t}}}\int_{V_0(\tilde{\mathbf{L}})} b^{C\,*}_{\tilde{\mathbf{t}}}(\tilde{\mathbf{r}})\,(\tilde{\mathbf{r}} - \tilde{\mathbf{L}})\,b^V_{\tilde{\mathbf{s}},\alpha}(\tilde{\mathbf{r}})\,d\,\tilde{\mathbf{r}}\exp(-\mathrm{i}\,\omega\,t), \quad (7)$$

where integration is taken over the volume V_0 of the unit cell at the node $\tilde{\mathbf{L}}$. Since Wannier functions are strongly localized within the respective unit cells at the nodes $\tilde{\mathbf{s}}$ and $\tilde{\mathbf{t}}$ the only option when the integral in (7) is not zero corresponds to $\tilde{\mathbf{s}} = \tilde{\mathbf{t}} = \tilde{\mathbf{L}}$ case. Thus, introducing the notation $\mathbf{p}_\alpha$ for the independent on a particular unit cell $\tilde{\mathbf{s}}$ transition dipole moment,

$$\mathbf{p}_\alpha = e\int_{V_0(\tilde{\mathbf{s}})} b^{C\,*}_{\tilde{\mathbf{s}}}(\tilde{\mathbf{r}})\,\tilde{\mathbf{r}}\,b^V_{\tilde{\mathbf{s}},\alpha}(\tilde{\mathbf{r}})\,d\,\tilde{\mathbf{r}} \ , \quad (8)$$

the following expression for the mean dipole moment $\mathbf{P}(\tilde{\mathbf{L}},t)$ of the lattice site $\tilde{\mathbf{L}}$ can be obtained:

$$\mathbf{P}(\tilde{\mathbf{L}},t) = \sum_\alpha C^{\alpha}_{\tilde{\mathbf{L}}\tilde{\mathbf{L}}}\,\mathbf{p}_\alpha\exp(-\mathrm{i}\omega t) \ , \quad (9)$$

i.e. $\mathbf{P}(\tilde{\mathbf{L}},t)$ is formed by the "node" dipole moments $\mathbf{p}_\alpha$ with weight coefficients $C^{\alpha}_{\tilde{\mathbf{L}}\tilde{\mathbf{L}}}$. By passing from discrete ($\tilde{\mathbf{L}}$) to continuous ($\tilde{\mathbf{r}}_d$) variable the discrete amplitude $C^{\alpha}_{\tilde{\mathbf{L}}\tilde{\mathbf{L}}}$ of electron-hole transition in the node $\tilde{\mathbf{L}}$ transforms into the exciton envelope wave function $\Psi^{\alpha}_{ex}(\tilde{\mathbf{r}}_d, \tilde{\mathbf{r}}_d)$. In the first approximation this function takes a form of the product of electron and hole wave functions of size-quantized states in QD, $\Psi^{\alpha}_{ex}(\tilde{\mathbf{r}}_d, \tilde{\mathbf{r}}_d) = \varphi_e(\tilde{\mathbf{r}}_d)\varphi^{\alpha}_h(\tilde{\mathbf{r}}_d)$ [7, 8]. Accordingly, under such passing to continuous variables the "node" dipole in the sum (9) transforms into the located at $\tilde{\mathbf{r}}_d$ "point" radiating dipole $\mathbf{p}_\alpha\exp(-\mathrm{i}\,\omega t)$, and by summing contributions of all QD unit cells into the total QD radiation the discrete sums of the type $\sum_{\tilde{\mathbf{L}}} C^{\alpha}_{\tilde{\mathbf{L}}\tilde{\mathbf{L}}} f(\tilde{\mathbf{L}})$ should be replaced by the integrals $\int \Psi^{\alpha}_{ex}(\tilde{\mathbf{r}}_d, \tilde{\mathbf{r}}_d) f(\tilde{\mathbf{r}}_d)\,d^3\tilde{r}_d$.

III. MODEL OF THE SYSTEM

Consider a system consisting of three mediums (figure 1).

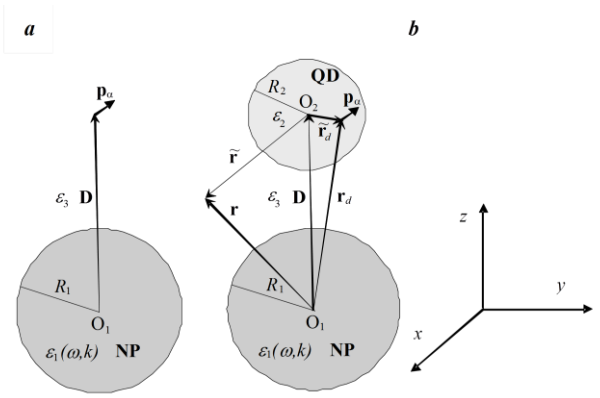

Fig 1. Point dipole in a vicinity of metal NP; structure has spherical symmetry (a). Semiconductor QD in a vicinity of metal NP; structure isn't spherically symmetric (b).

1) *Spherical metal NP* of the radius R_1 characterized by a non-local permittivity $\varepsilon_1(\omega,k)$. Using non-local permittivity instead of local one $\varepsilon_1(\omega)$, as well as account for additional electron scattering on the NP walls allows calculations for NP of extremely small sizes (less than 10 nm in diameter) by the methods of classical macroscopic electrodynamics [9]. In line with [9-11] in optical range of frequencies $\varepsilon_1(\omega,k)$ can be expressed as:

$$\varepsilon_1^{T(L)}(\omega,k)=1+\varepsilon_{ib}(\omega)+\varepsilon_{pl}^{T(L)}(\omega,k)\ , \qquad (10)$$

where second term on the right-hand side is due to interband electronic transitions (the contribution of bound electrons), and the third term,

$$\varepsilon_{pl}^{T(L)}(\omega,k)=-\omega_p^2\Big/\left\{\omega\left[\omega+\mathrm{i}\left(\Gamma+Av_F/R_1\right)\right]-\beta_{T(L)}^2k^2\right\}\ , \qquad (11)$$

is due to electronic transitions within the conduction band (the contribution of free electrons). Indices *T* and *L* are used to denote transverse (*T*) and longitudinal (*L*) components of the permittivity. Within the hydrodynamic model $\beta_L=\sqrt{3/5}\,v_F$, where v_F is the Fermi velocity ($v_F\approx 1.4\cdot 10^8$ cm/s in the case of gold or silver), while β_T equals zero and therefore $\varepsilon_1^T(\omega,k)\equiv\varepsilon_1^T(\omega,0)$. The decay constant of the bulk material $\Gamma\approx v_F/l_f$, where l_f is the mean free path of electrons, constant *A* takes values in the $0.1\div 0.7$ range [9] depending on the mechanism of electron scattering on the NP walls and other factors, $\omega_p^2=4\pi ne^2/m^*$ is the plasma frequency of the NP material. At a given frequency ω the wave number k_T of the transverse electromagnetic waves in the NP could be determined from the condition $k_T^2=\varepsilon_1^T(\omega,0)\,\omega^2/c^2$, while the wave number k_L of the longitudinal electric oscillations from the condition $\varepsilon_1^L(\omega,k_L)=0$.

2) *Spherical semiconductor QD* of the radius R_2 with background permittivity ε_2. The distance between QD and NP centers is $D>R_1+R_2$.

3) *Barrier matrix* with permittivity ε_3.

The formulated electrodynamic problem for the oscillating point dipole $\mathbf{p}_\alpha$ inside QD is considered below using Jackson formalism [12], where vector spherical harmonics $\mathbf{X}_{lm}(\Omega)$ are used in multipole expansions of transverse electric and magnetic fields. As for the calculation of electromagnetic fields in QD+NP system the boundary conditions on both semiconductor QD and metal NP spherical surfaces should be satisfied, the main problem becomes the establishing a connection between the multipole components of the fields in two different spherical coordinate system with the centers O_1 in metal NP and O_2 in semiconductor QD.

IV. RELATIONSHIP BETWEEN MULTIPOLE EXPANSIONS OF ELECTROMAGNETIC FIELD IN TWO COORDINATE SYSTEMS

By calculating electromagnetic fields in nonspherical QD+NP system the best solution would be to use separately spherical symmetry of QD and NP. According to [12] in the *n*-th material with permittivity ε_n the electric and magnetic fields in spherically symmetric cases can be expressed as following multipole expansions :

$$\mathbf{E}(\mathbf{r})=\sum_{l=1}^{\infty}\sum_{m=-l}^{l}\left\{\frac{\mathrm{i}}{k_0\,\varepsilon_\mathrm{n}}a_E(l,m)\,\mathrm{curl}\left[f_l(k_\mathrm{n}r)\,\mathbf{X}_{lm}(\Omega)\right]+\right.\ , \qquad (12)$$
$$\left.+a_M(l,m)\,f_l(k_\mathrm{n}r)\mathbf{X}_{lm}(\Omega)\right\}$$

$$\mathbf{B}(\mathbf{r})=\sum_{l=1}^{\infty}\sum_{m=-l}^{l}\left\{a_E(l,m)\,f_l(k_\mathrm{n}r)\mathbf{X}_{lm}(\Omega)-\right.\ , \qquad (13)$$
$$\left.-\frac{\mathrm{i}}{k_0}a_M(l,m)\,\mathrm{curl}\left[f_l(k_\mathrm{n}r)\,\mathbf{X}_{lm}(\Omega)\right]\right\}$$

where $k_0=\omega/c$, $k_\mathrm{n}=k_0\sqrt{\varepsilon_\mathrm{n}}$, $a_E(l,m)$ and $a_M(l,m)$ are the multipole amplitudes (coefficients) of the electric and magnetic types, respectively, $f_l(x)$ is the spherical Bessel function $j_l(x)$ or the first kind Hankel function $h_l^{(1)}(x)$ (hereafter denoted $h_l(x)$ as well) or their combination depending on the region of space and asymptotic or boundary conditions that should be provided. Normalized vector spherical harmonics $\mathbf{X}_{lm}(\Omega)$ (symbol Ω means angular variables $\{\theta,\varphi\}$) are expressed as follows:

$$\mathbf{X}_{lm}=\frac{r}{\mathrm{i}\sqrt{l(l+1)}}\left[\mathbf{e}_r\times\nabla Y_{lm}\right]=\ , \qquad (14)$$
$$=\frac{1}{\mathrm{i}\sqrt{l(l+1)}}\left[-\mathbf{e}_\theta\frac{1}{\sin\theta}\frac{\partial}{\partial\varphi}+\mathbf{e}_\varphi\frac{\partial}{\partial\theta}\right]Y_{lm}$$

$Y_{lm}(\Omega)=\left[(2l+1)(l-m)!/(4\pi(l+m)!)\right]^{1/2}P_l^m(\cos\theta)\exp(\mathrm{i}m\varphi)$ are the normalized scalar spherical harmonics, $P_l^m(x)$ are the associated Legendre functions. Accordingly,

$$\mathrm{curl}\left[f_l(k_\mathrm{n}r)\,\mathbf{X}_{lm}(\Omega)\right]=\mathrm{i}\left\{\sqrt{l(l+1)}\,\frac{f_l(k_\mathrm{n}r)}{r}\,Y_{lm}(\Omega)\,\mathbf{e}_r+\right.\ . \qquad (15)$$
$$\left.+\frac{1}{\sqrt{l(l+1)}}\frac{d\left(rf_l(k_\mathrm{n}r)\right)}{dr}\nabla Y_{lm}(\Omega)\right\}$$

In (14) and (15) $\mathbf{e}_r$,$\mathbf{e}_\theta$ and $\mathbf{e}_\varphi$ are the orts of spherical coordinate system. The following orthogonality and normalization condition are used hereafter:

$$\int\mathbf{X}_{l'm'}^*(\Omega)\cdot\mathbf{X}_{lm}(\Omega)\,d\Omega=\delta_{ll'}\delta_{mm'}\ , \qquad (16)$$

$$\int \mathrm{curl}[\mathbf{X}^*_{l'm'}(\Omega)]\cdot\mathrm{curl}[\mathbf{X}_{lm}(\Omega)]\ d\Omega=\delta_{ll'}\delta_{mm'}\frac{l(l+1)+1}{r^2}\ , \quad (17)$$

$$\int \nabla Y^*_{l'm'}(\Omega)\cdot\nabla Y_{lm}(\Omega)\ d\Omega=l(l+1)/r^2\,\delta_{ll'}\delta_{mm'}\ , \quad (18)$$

$$\int \mathbf{X}^*_{l'm'}(\Omega)\cdot\mathrm{curl}[\mathbf{X}_{lm}(\Omega)]\ d\Omega=0\ , \quad (19)$$

$$\int \mathbf{X}^*_{l'm'}(\Omega)\cdot\nabla Y_{lm}(\Omega)\ d\Omega=0\ , \quad (20)$$

$$\int Y^*_{l'm'}(\Omega)\,Y_{lm}(\Omega)\,d\Omega=\delta_{ll'}\delta_{mm'}\ . \quad (21)$$

It could be found in line with [12], that for the located at point $\mathbf{r}_d$ oscillating dipole $p_\alpha\,\mathbf{n}_\alpha\exp(-\mathrm{i}\omega t)$ ($\mathbf{n}_\alpha$ is a unit vector along the dipole direction) the multipole coefficients $a_E(l,m)$ and $a_M(l,m)$ in expansions (12) and (13) should take the following form:

$$a_{E,d}(l,m)=4\pi\,p_\alpha\,k_\mathrm{n}k_0\,\mathbf{n}_\alpha\cdot\mathrm{curl}_{\mathbf{r}_d}[g_l(k_\mathrm{n}r_d)\mathbf{X}^*_{lm}(\Omega_d)]\ , \quad (22)$$

$$a_{M,d}(l,m)=4\pi\,\mathrm{i}\,p_\alpha\,k_\mathrm{n}k_0^2\,g_l(k_\mathrm{n}r_d)\,\mathbf{n}_\alpha\cdot\mathbf{X}^*_{lm}(\Omega_d)\ , \quad (23)$$

and there is the following correlation between functions $f_l(k_\mathrm{n}r)$ in (12) and (13) and $g_l(k_\mathrm{n}r_d)$ in (22) and (23):

$$f_l(k_\mathrm{n}r)=j_l(k_\mathrm{n}r)\,,\ g_l(k_\mathrm{n}r_d)=h_l(k_\mathrm{n}r_d)\quad\text{if}\quad r<r_d\ , \quad (24)$$

$$f_l(k_\mathrm{n}r)=h_l(k_\mathrm{n}r)\,,\ g_l(k_\mathrm{n}r_d)=j_l(k_\mathrm{n}r_d)\quad\text{if}\quad r>r_d\ . \quad (25)$$

For the convenience of further consideration following notations are introduced:

$$a^{>}_{E,d}(l,m)=4\pi\,p_\alpha\,k_\mathrm{n}k_0\,\mathbf{n}_\alpha\cdot\mathrm{curl}_{\mathbf{r}_d}[j_l(k_\mathrm{n}r_d)\mathbf{X}^*_{lm}(\Omega_d)]\ , \quad (26)$$

$$a^{<}_{E,d}(l,m)=4\pi\,p_\alpha\,k_\mathrm{n}k_0\,\mathbf{n}_\alpha\cdot\mathrm{curl}_{\mathbf{r}_d}[h_l(k_\mathrm{n}r_d)\mathbf{X}^*_{lm}(\Omega_d)]\ , \quad (27)$$

$$a^{>}_{M,d}(l,m)=4\pi\,\mathrm{i}\,p_\alpha\,k_\mathrm{n}k_0^2\ j_l(k_\mathrm{n}r_d)\,\mathbf{n}_\alpha\cdot\mathbf{X}^*_{lm}(\Omega_d)\ , \quad (28)$$

$$a^{<}_{M,d}(l,m)=4\pi\,\mathrm{i}\,p_\alpha\,k_\mathrm{n}k_0^2\ h_l(k_\mathrm{n}r_d)\,\mathbf{n}_\alpha\cdot\mathbf{X}^*_{lm}(\Omega_d)\ . \quad (29)$$

IV.I. Homogeneous medium

To find relations between the coefficients of electromagnetic field multipole expansions in two spherical coordinate systems with the centers in points O_1 and O_2 (see figure 1) consider for the first the case of a homogeneous medium with $\varepsilon_1=\varepsilon_2=\varepsilon_3$. According to (12), in these two systems multipole expansions of point dipole electric field should have the following form:

$$\mathbf{E}(\mathbf{r})=\sum_{l=1}^{\infty}\sum_{m=-l}^{l}\left\{\frac{\mathrm{i}}{k_0\,\varepsilon_3}a_E(l,m)\,\mathrm{curl}\big[\,f_l(k_3r)\,\mathbf{X}_{lm}(\Omega)\big]+\right.$$
$$\left.+a_M(l,m)\,f_l(k_3r)\mathbf{X}_{lm}(\Omega)\ \right\}\ , \quad (30)$$

$$\mathbf{E}(\tilde{\mathbf{r}})=\sum_{l=1}^{\infty}\sum_{m=-l}^{l}\left\{\frac{\mathrm{i}}{k_0\,\varepsilon_3}\tilde{a}_{E,d}(l,m)\,\mathrm{curl}_{\tilde{\mathbf{r}}}\big[\,f_l(k_3\tilde{r})\,\mathbf{X}_{lm}(\tilde{\Omega})\big]+\right.$$
$$\left.+\ \tilde{a}_{M,d}(l,m)\,f_l(k_3\tilde{r})\mathbf{X}_{lm}(\tilde{\Omega})\right\}\ . \quad (31)$$

For the field is independent on a particular coordinate system used for its presentation $\mathbf{E}(\mathbf{r})\equiv\mathbf{E}(\tilde{\mathbf{r}})$ in every point of the space. Multiplying this identity on $\mathbf{X}^*_{l'm'}(\Omega)$ and integrating over solid angle Ω with an account for (16) and (19) the following connection between multipole coefficient a_M and multipole coefficients $\tilde{a}^{>}_{M,d}$ and $\tilde{a}^{>}_{E,d}$ for the region $\tilde{r}>\tilde{r}_d$ can be established:

$$a_M(l,m)=\sum_{l'=1}^{\infty}R^m_{ll'}\left[\,F^m_{ll'}(D)\,\tilde{a}^{>}_{M,d}(l',m)\ +\right.$$
$$\left.+\frac{m(k_3D)}{l(l+1)\sqrt{\varepsilon_3}}G^m_{ll'}(D)\,\tilde{a}^{>}_{E,d}(l',m)\right]\ . \quad (32)$$

Analogously, from the identity of magnetic fields $\mathbf{B}(\mathbf{r})\equiv\mathbf{B}(\tilde{\mathbf{r}})$ we have

$$a_E(l,m)=\sum_{l'=1}^{\infty}R^m_{ll'}\left[\,F^m_{ll'}(D)\,\tilde{a}^{>}_{E,d}(l',m)\ -\right.$$
$$\left.-\frac{m(k_3D)\sqrt{\varepsilon_3}}{l(l+1)}G^m_{ll'}(D)\,\tilde{a}^{>}_{M,d}(l',m)\right]\ . \quad (33)$$

In (32) and (33)

$$R^m_{ll'}=\frac{\sqrt{(2l+1)(2l'+1)}}{2}\sqrt{\frac{l\ (l+1)}{l'(l'+1)}\,\frac{(l-m)!}{(l+m)!}\,\frac{(l'-m)!}{(l'+m)!}}\ , \quad (34)$$

$$F^m_{ll'}(D)=\frac{1}{f_l(k_3r)}\left\{\int_{-1}^{1}P^m_l(x)\left[\frac{l'(l'+1)}{l(l+1)}\,\frac{f_{l'}(k_3\tilde{r})}{s^2}\left(1-x\frac{D}{r}\right)P^m_{l'}(\tilde{x})+\right.\right.$$
$$\left.\left.+\frac{(k_3D)}{l(l+1)}\left(\frac{f_{l'}(k_3\tilde{r})}{k_3\tilde{r}}+f'_{l'}(k_3\tilde{r})\right)(1-\tilde{x}^2)\frac{dP^m_{l'}(\tilde{x})}{d\,\tilde{x}}\right]dx\right\}\ , \quad (35)$$

$$G^m_{ll'}(D)=[1/\,f_l(k_3r)]\int_{-1}^{1}f_{l'}(k_3\tilde{r})\,P^m_l(x)\,P^m_{l'}(\tilde{x})\,dx \quad (36)$$

where $f'(y)$ means $df(y)/dy$, $\tilde{x}=(x-D/r)/s$, $\tilde{r}=rs$, $s=[\,1+(\,D/r\,)^2\ -2\,xD/r\,]^{1/2}$. Formulae (32)-(36) have been obtained accounting for the following expressions for the orts $\mathbf{e}_{\tilde{r}}$, $\mathbf{e}_{\tilde{\theta}}$ and $\mathbf{e}_{\tilde{\varphi}}$:

$$\mathbf{e}_{\tilde{r}}=[(1-xD/r)\mathbf{e}_r+\sqrt{1-x^2}\,(D/r)\mathbf{e}_\theta]/s\ , \quad (37)$$

$$\mathbf{e}_{\tilde{\theta}}=[-\sqrt{1-x^2}\,(D/r)\mathbf{e}_r+(1-xD/r)\mathbf{e}_\theta]/s\ , \quad (38)$$

$$\mathbf{e}_{\tilde{\varphi}}=\mathbf{e}_\varphi\ . \quad (39)$$

In accordance with explicit expressions (35) and (36) the connection coefficients $F^m_{ll'}$ and $G^m_{ll'}$ look like coordinate-dependent ones. However, their independence on the "connection sphere" radius r can be easily verified by making numerical integrations in (35) and (36). In the case of $\tilde{r}>\tilde{r}_d$ function $f_{l'}(k_3\tilde{r})$ in the integrands (35) and (36) is the spherical Hankel function of the first kind $h^{(1)}_{l'}(k_3\tilde{r})$. Function $f_l(k_3r)$ is the spherical Bessel function $j_l(k_3r)$ in the region $r<r_d$ and spherical Hankel function $h^{(1)}_l(k_3r)$ in the region $r>r_d$. Consider the case when dipole is located near O_2 center ($\tilde{r}_d\to 0$). Then $f_l(k_3r)=j_l(k_3r)$ at $r<D$ and $f_l(k_3r)=h^{(1)}_l(k_3r)$ at $r>D$. As an example, the results of numerical calculations of F^m_{Ll} and G^m_{Ll} are shown in figures 2 and 3 for the set of parameters {L,l,m} = {3,4,2}, $\varepsilon_3=3$, $\lambda_0=500$ nm, $D=5$ nm. As seen from these figures, connection coefficients $F^m_{ll'}$ and $G^m_{ll'}$ really do not depend on r, i.e. they are the constants. At arbitrary finite $\tilde{r}_d$ the parts of the dependencies in the region $D-\tilde{r}_d<r<D+\tilde{r}_d$ should be deleted in these figures.

IV.II. Semiconductor QD in barrier matrix

Consider radiation of a point dipole located inside QD with dielectric constant ε_2, the QD being formed in barrier matrix with dielectric constant ε_3. The field, which is incident on the inner surface of the QD, reflected inside QD field and the field transmitted into barrier matrix could be written in the form of (12) and (13) with corresponding spherical Bessel or Hankel functions depending on the necessity to provide the field finiteness

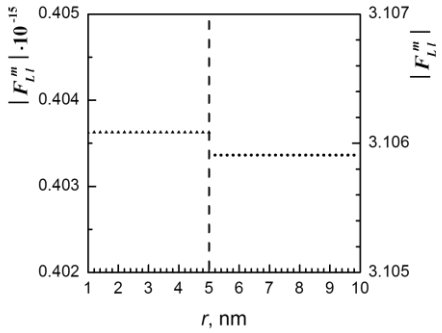

Fig. 2. Connection coefficient $|F_{Ll}^m|$ as function of r in the case of L=3, l=4, m=2, D = 5 nm. In the $r < D$ region $f_L(k_3r) \equiv j_L(k_3r)$, values $|F_{Ll}^m|$ refer to the left axis of ordinates. In the $r > D$ region $f_L(k_3r) \equiv h_L(k_3r)$, values $|F_{Ll}^m|$ refer to the right axis of ordinates.

at $\tilde{r} = 0$ or right asymptotic behavior at $\tilde{r} \to \infty$. In a standard way [12], using (16)-(21) two systems of

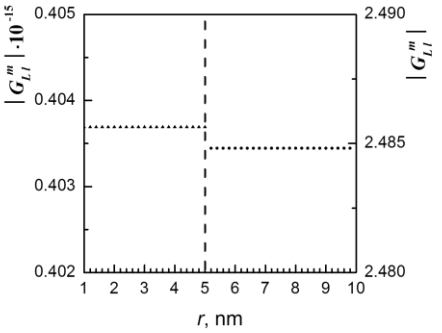

Fig. 3. Connection coefficient $|G_{Ll}^m|$ as function of r for the same parameters and conditions as in Fig. 2.

algebraic equations for the multipole coefficients could be obtained from the continuity conditions for the tangential components of the electric and magnetic fields at the QD surface. As follows from these equations the relationships between the multipole coefficients $\tilde{a}_M^{(t)}(l,m)$ and $\tilde{a}_E^{(t)}(l,m)$ of electromagnetic field penetrated into the barrier matrix and corresponding coefficients $\tilde{a}_{M,d}^{>}(l,m)$ and $\tilde{a}_{E,d}^{>}(l,m)$ of the point dipole electromagnetic field incident on the QD inner surface have the following form:

$$\tilde{a}_M^{(t)}(l,m) = V_{23}^M(l)\, \tilde{a}_{M,d}^{>}(l,m)\ , \tag{40}$$

$$\tilde{a}_E^{(t)}(l,m) = V_{23}^E(l)\, \tilde{a}_{E,d}^{>}(l,m)\ , \tag{41}$$

$$V_{23}^M(l) = \frac{\mathrm{i}\sqrt{\varepsilon_2}}{(k_2R_2)^2} \frac{1}{\sqrt{\varepsilon_3}\, j_l(k_2R_2)\, h_l'(k_3R_2) - \sqrt{\varepsilon_2}\, h_l(k_3R_2)\, j_l'(k_2R_2)} \tag{42}$$

$$V_{23}^E(l) = \frac{\mathrm{i}\varepsilon_3}{k_2R_2} \frac{1}{\varepsilon_2\, j_l(k_2R_2)\, [xh_l(x)]'_{x=k_3R_2} - \varepsilon_3\, h_l(k_3R_2)\, [x\, j_l(x)]'_{x=k_2R_2}} \tag{43}$$

The connection between multipole coefficients $a_M^{(i)}$ and $a_E^{(i)}$ of electromagnetic field outside QD written in coordinate system O_1 and multipole coefficients $\tilde{a}_M^{(t)}$ and $\tilde{a}_E^{(t)}$ of the same field written in coordinate system O_2 is analogous to the homogeneous medium case:

$$a_M^{(i)}(l,m) = \sum_{l'=1}^{\infty} R_{ll'}^m \left[F_{ll'}^m(D)\, \tilde{a}_M^{(t)}(l',m) + \frac{m(k_3D)}{l(l+1)\sqrt{\varepsilon_3}} G_{ll'}^m(D)\, \tilde{a}_E^{(t)}(l',m) \right], \tag{44}$$

$$a_E^{(i)}(l,m) = \sum_{l'=1}^{\infty} R_{ll'}^m \left[F_{ll'}^m(D)\, \tilde{a}_E^{(t)}(l',m) - \frac{m(k_3D)\sqrt{\varepsilon_3}}{l(l+1)} G_{ll'}^m(D)\, \tilde{a}_M^{(t)}(l',m) \right]. \tag{45}$$

V. INTERACTION OF RADIATION WITH METAL NP

If spherical metal NP with the center at O_1 (see figure 1,b) is additionally placed into the above considered system, then electromagnetic field radiated by QD point dipole scatters at the NP. In previous section the multipole coefficients $a_E^{(i)}$ and $a_M^{(i)}$ of the field radiated by QD in the coordinate system with the center at O_1 have already been determined. This field can be considered now as being incident on the NP:

$$\boldsymbol{E}_3^{(i)}(\boldsymbol{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^{l} \left\{ \frac{\mathrm{i}}{k_0\, \varepsilon_3} a_E^{(i)}(l,m)\ \mathrm{curl}[\, j_l(k_3r)\, \boldsymbol{X}_{lm}(\Omega)\,] + a_M^{(i)}(l,m)\, j_l(k_3r)\, \boldsymbol{X}_{lm}(\Omega) \right\}, \tag{46}$$

$$\boldsymbol{B}_3^{(i)}(\boldsymbol{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^{l} \left\{ a_E^{(i)}(l,m)\, j_l(k_3r)\, \boldsymbol{X}_{lm}(\Omega) - \frac{\mathrm{i}}{k_0} a_M^{(i)}(l,m)\ \mathrm{curl}[\, j_l(k_3r)\, \boldsymbol{X}_{lm}(\Omega)] \right\}. \tag{47}$$

Scattered (reflected) field can be expressed by the following formulae:

$$\boldsymbol{E}_3^{(r)}(\boldsymbol{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^{l} \left\{ \frac{\mathrm{i}}{k_0\, \varepsilon_3} a_E^{(r)}(l,m)\ \mathrm{curl}[\, h_l(k_3r)\, \boldsymbol{X}_{lm}(\Omega)\,] + a_M^{(r)}(l,m)\, h_l(k_3r)\, \boldsymbol{X}_{lm}(\Omega) \right\}, \tag{48}$$

$$\boldsymbol{B}_3^{(r)}(\boldsymbol{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^{l} \left\{ a_E^{(r)}(l,m)\, h_l(k_3r)\, \boldsymbol{X}_{lm}(\Omega) - \frac{\mathrm{i}}{k_0} a_M^{(r)}(l,m)\ \mathrm{curl}[\, h_l(k_3r)\, \boldsymbol{X}_{lm}(\Omega)] \right\}. \tag{49}$$

Electric field $\mathbf{E}_1^{(t)}(\mathbf{r})$ induced (transmitted) in metal NP is the sum of the transverse and longitudinal fields, $\mathbf{E}_1^{(t)}(\mathbf{r}) = \mathbf{E}_1^T(\mathbf{r}) + \mathbf{E}_1^L(\mathbf{r})$, where the transverse electric field can be written as

$$\mathbf{E}_1^T(\mathbf{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^{l} \left\{ \frac{\mathrm{i}}{k_0\, \varepsilon_1^T(\omega,0)} a_E^{(t)}(l,m)\ \mathrm{curl}[\, j_l(k_Tr)\, \mathbf{X}_{lm}(\Omega)] + a_M^{(t)}(l,m)\, j_l(k_Tr)\, \mathbf{X}_{lm}(\Omega) \right\} \tag{50}$$

and longitudinal electric field that arises in metal NP due to spatial dispersion (see (10) and (11)), respectively as

$$\mathbf{E}_1^L(\mathbf{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^{l} a_L(l,m) \frac{1}{k_L} \nabla[\, j_l(k_L\, r) Y_{lm}(\Omega)]\ . \tag{51}$$

The magnetic field inside the metal NP can be expressed as follows:

$$\mathbf{B}_1^{(t)}(\mathbf{r}) = \sum_{l=1}^{\infty}\sum_{m=-l}^{l}\left\{ a_E^{(t)}(l,m)\, j_l(k_T r)\,\mathbf{X}_{lm}(\Omega) - \right. \quad . \quad (52)$$
$$\left. - \frac{\mathrm{i}}{k_0} a_M^{(t)}(l,m)\ \mathrm{curl}[\, j_l(k_T r)\,\mathbf{X}_{lm}(\Omega)] \right\}$$

From the continuity conditions for the tangential components of electric and magnetic fields on the NP surface and additional boundary condition in the form of turning to zero of the normal component of free carrier's current density on the NP surface [10, 11], the following expressions for the coefficients of multipole expansions (48)-(51) could be found using (16)-(19):

$$a_M^{(r)}(l,m) = V_{33}^M(l)\, a_M^{(i)}(l,m)\ , \quad (53)$$

$$a_E^{(r)}(l,m) = V_{33}^E(l)\, a_E^{(i)}(l,m)\ , \quad (54)$$

$$a_M^{(t)}(l,m) = V_{31}^M(l)\, a_M^{(i)}(l,m)\ , \quad (55)$$

$$a_E^{(t)}(l,m) = V_{31}^E(l)\, a_E^{(i)}(l,m)\ , \quad (56)$$

$$a_L(l,m) = V_{31}^L(l)\, a_E^{(i)}(l,m)\ , \quad (57)$$

$$V_{33}^M(l) = \left[\sqrt{\varepsilon_3}\ j_l(k_T R_1)\, j_l'(k_3 R_1) - \right. \quad , \quad (58)$$
$$\left. - \sqrt{\varepsilon_1^T(\omega,0)}\ j_l(k_3 R_1)\, j_l'(k_T R_1) \right] \Big/ \Delta_M$$

$$V_{33}^E(l) = \left\{ \varepsilon_3\, j_l(k_3 R_1)[x j_l(x)]'_{x=k_T R_1} + (C\varepsilon_1^T(\omega,0) - \varepsilon_3)\, A - \right. \quad , \quad (59)$$
$$\left. - \varepsilon_1^T(\omega,0)\, j_l(k_T R_1)[x j_l(x)]'_{x=k_3 R_1} \right\} \Big/ \Delta_{EL}$$

$$V_{31}^M(l) = -\mathrm{i}\sqrt{\varepsilon_3} \Big/ \left[(k_3 R_1)^2 \Delta_M \right], \quad (60)$$

$$V_{31}^E(l) = \mathrm{i}\varepsilon_1^T(\omega,0) \Big/ (k_3 R_1\, \Delta_{EL}), \quad (61)$$

$$V_{31}^L(l) = -\frac{i\sqrt{l(l+1)}\left(C\varepsilon_1^T(\omega,0) - \varepsilon_3\right) j_l(k_T R_1)}{\varepsilon_3 k_3 k_0 R_1^2\, j_l'(k_L R_1)\Delta_{EL}}\ , \quad (62)$$

$$\Delta_M = \sqrt{\varepsilon_1^T(\omega,0)}\ h_l(k_3 R_1)\, j_l'(k_T R_1) - \quad (63)$$
$$- \sqrt{\varepsilon_3}\ j_l(k_T R_1)\, h_l'(k_3 R_1)$$

$$\Delta_{EL} = \varepsilon_1^T(\omega,0)\, j_l(k_T R_1)[x h_l(x)]'_{x=k_3 R_1} - \quad , \quad (64)$$
$$- \varepsilon_3 h_l(k_3 R_1)[x j_l(x)]'_{x=k_T R_1} - (C\varepsilon_1^T(\omega,0) - \varepsilon_3)\, B$$

$$A = l(l+1)\, j_l(k_L R_1)\, j_l(k_T R_1)\, j_l(k_3 R_1) \Big/ \left[k_L R_1\, j_l'(k_L R_1) \right], \quad (65)$$

$$B = l(l+1)\, j_l(k_L R_1)\, j_l(k_T R_1)\, h_l(k_3 R_1) \Big/ \left[k_L R_1\, j_l'(k_L R_1) \right], \quad (66)$$

$$C = \varepsilon_3 / [1 + \varepsilon_{ib}(\omega)]\ . \quad (67)$$

VI. CONCLUSION

Thus, in this paper all electromagnetic fields in the nonspherical "semiconductor QD + metal NP" structure are consistently calculated in the single scattering approximation for the case of radiation of a point dipole located inside QD. It enables finding the power and the rate of radiation as well as the intensity of electromagnetic energy absorption and the rate of nonradiative losses in the system under consideration. Hence the efficiency of radiation depending on the distance D between QD and NP, radii R_1 and R_2 and other parameters as well as its
change relatively to the case of isolated QD could be calculated. After summing the contributions from all QD unit cells in line with section II analogous characteristics including luminescence efficiency change could be found for semiconductor QD as a whole. Using found connections between multipole coefficients of the electromagnetic field in two different spherical coordinate systems with the centers in QD and metal NP the developed theory could be generalized to account for multiple scatterings of electromagnetic field between QD and NP.

ACKNOWLEDGMENTS

The work was done as part of a complex program of basic researches "Fundamental Problems of nanostructured systems, nanomaterials and nanotechnologies" NAS of Ukraine (Theme # 52).

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