

Inter*net* Electronic Journal Nanociencia et Moletrónica

Octubre 2004, Vol. 2; N°2, págs. 302-310

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recibido: 28 de septiembre de 2004

revisado: octubre 2004

publicado: octubre 2004

Citation of the article:

G. N. Burlak¹, O. Starostenko², A. Zamudio-Lara¹, P. A. Márquez Aguilar¹ "The Electromagnetic Field Quantization in Microspherical Structures", Internet Electrón. J. Nanocs. Moletrón. 2004, 2(2), 300-309

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Abstract

A simple approach to the electromagnetic field quantization in nonuniform spherical nano-structures is proposed. It allows one to obtain the vacuum field amplitude in general form. We found that such a quantity strongly depends on the spherical quantum number.

1 Introduction

The phenomenon of modification of the spectrum of spontaneous emission (SE) in cavities in comparison with a vacuum case is studied for a rather long time and now is well understood [1]-[9]. Recent progress in constructing certain types of microcavities such as microspheres has rendered to make clearer the specific quantum properties of field in such microobjects. Last years in a number of works [10]-[12], were observed the SE in microspheres with sizes about $5-20\mu m$. The coupling of electronic and photonic states is demonstrated for a single photonic dot by the observation of whispering gallery modes (WGM) in the spectrum of SE of the embedded *CdSe* quantum dots.

In[13] the estimations of the optimum sizes of microspheres for effective atoms-field interaction in WGM regimes are done. The WGMs of quartz microspheres are investigated for the purpose of strong coupling between single photons and atoms. Other important direction is studying the SE from photonic crystals [14]-[16]. Modern period is characterized by further reduction of spatial scales and transition to the nanostructures. In [17] the spontaneous decay process of an excited atom placed inside or outside a carbon nanotube is analyzed and sufficient increasing the atomic spontaneous decay rate is predicted. In [18] was measured fluorescence lifetimes of emitters embedded in isolated single dielectric nanospheres. The results show inhibition of the SE up to 3 times. Such migration to the nano-scales and nanospheres arises the situation when the low frequencies range of WGM spectrum become higher than the typical frequencies of the atoms transition. The interaction becomes resonant for the photons modes, having the smaller spherical quantum numbers. Therefore in spherical nanostructures one can expect the new peculiarities of the fundamental atom-field interactions.

The procedure of quantization of the electromagnetic field is well known [8], [9]. In a planewaves case the following expression for an electric field turns out $E = \mathbf{E}_0(a^+e^{i\theta} + ae^{-i\theta})$, where the quantity $\mathbf{E}_0 = (\hbar\omega/2\varepsilon_0 V)^{1/2}$ has the dimensions of an electric field, V is a volume, $\theta = \omega t - \vec{k}\vec{r}$, a^+ and a are creation and annihilation operators of photon on a corresponding mode respectively. Such a field is included into the atom-field interactions part of Schrödinger equation $H_{af} = -e(\vec{r} \cdot \vec{E})$. For exited atom this interaction occurs even for vacuum state $|0\rangle$ and can stimulate the atom to emit spontaneously. It is well-known that the expecting value of linear polarized field vanish $\langle n|E|n\rangle = 0$, but $\langle n|E^2|n\rangle = E_0^2(2n+1)$ for a number state $|n\rangle$ [9]. The nonzero fluctuations occur even for a vacuum state $|0\rangle$ i.e. $\langle n|E^2|n\rangle = E_0^2$. From the latter the fundamental quantity E_0 as the field's vacuum amplitude (or field per photon) can be recognized. In this paper a simple approach to the fields' quantization in the nonuniform spherical structures from first principles is proposed. We show, that simple a plane-waves behavior:

 $(\mathbf{E}_0)^2 \sim \omega^{1/2}$ is modified in microspheres. Such peculiarity becomes important due to a recent study the fundamental effects of field-atom coupling in microspheres [10], [11], [19] and ensembles of microspheres [20].

2 Description of solution

We study a spherical multilayered structure placed in a quantized sphere radius a_0 (see Fig.1). Due to the spherical geometry the state of electromagnetic field in microsphere is characterized with indexes ν , m and j (radial, spherical and azimuthal quantum number) accordingly. In this case for a photon state we write $|n_{\tilde{k}}\rangle \rightarrow |n_{\nu m}\rangle$, $\omega_{\tilde{k}} \rightarrow \omega_{\nu m}$ (the eigenfrequencies of sphere do not depend on azimuthal quantum number [21]), $E_{\tilde{k}} \rightarrow E_{\nu m j}$.



Fig.1. Geometry of system.

The electric and magnetic fields \vec{E}, \vec{H} in spherical polar coordinates (r, θ, φ) have form

$$\vec{E}(r,\theta,\varphi) = \sum_{\nu,m,j} \vec{E}_{\nu m j}(r,\theta,\varphi), \quad \vec{H}(r,\theta,\varphi) = \sqrt{\frac{\varepsilon_0}{\mu_0}} \sum_{\nu,m,j} \vec{H}_{\nu m j}(r,\theta,\varphi), \quad (1)$$

$$\vec{E}_{vmj}(r,\theta,\varphi) = A_{vm} \sum_{s} \varepsilon_{s}(r,\theta,\varphi) \hat{e}_{s}, \vec{H}_{vmj}(r,\theta,\varphi) = A_{vm} \sum_{s} h_{s}(r,\theta,\varphi) \hat{e}_{s}$$
(2)

where $A_{vm} = A_{0vm}e^{-i\omega t}$, A_{0vm} is a complex amplitude, $s = r, \theta, \varphi$, and $\hat{e}_{r,\theta,\varphi}$ are the basis set for spherical coordinates.

Due to the modes orthogonality we can study the modes oscillations separately.

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In this stage we omit the indexes v, m, j for notational simplicity. Two different polarization, TM and TE, are allowed. In TM case $h_r = 0$, while for TE case $e_r = 0$. We start from the TM wave case. The field's components in (1), (2) one can write in the next form

$$\varepsilon_s(y,\theta,\varphi) = AF_s, h_s(y,\theta,\varphi) = iAf_s,$$
(3)

$$F_r = \frac{m(m+1)}{k_0^2 r^2 \varepsilon} \Pi, \ F_\theta = \frac{1}{k_0^2 r \varepsilon} \frac{\partial^2 \Pi}{\partial r \partial \theta}, \ F_\varphi = \frac{1}{k_0^2 r \varepsilon \sin \theta} \frac{\partial^2 \Pi}{\partial r \partial \varphi},$$
(4)

$$f_r = 0, \ f_\theta = -\frac{1}{k_0 r \sin \theta} \frac{\partial \Pi}{\partial \varphi}, \ f_\varphi = \frac{1}{k_0 r} \frac{\partial \Pi}{\partial \theta}, \tag{5}$$

where $\Pi(r,\theta,\varphi)$ is Debye potential, which is given by $\Pi(r,\theta,\varphi) = R_m(r)Y_m^j(\theta,\varphi)$, $Y_m^j(\theta,\varphi)$ are spherical functions, $k_0 = \omega/c$ and the radial part $R_m(r)$ for *TM* waves obeys the next equation $\varepsilon \frac{d}{dr} [\frac{1}{\varepsilon} \frac{dR_m}{dr}] + [\varepsilon - \frac{m(m+1)}{r^2}]R_m = 0$. In inhomogeneous microspheres the dielectric permittivity is $\varepsilon = \varepsilon(r)$. Since the equations for *TE* fields can be received by similar way [Stratt41], we do not write down corresponding formulas separately. Equations (1)-(5) contain all the necessary physical information regarding the field's quantization. For brevity we omit some details of general algebra from the canonical theory, which is known quite well [8], [9]. Our purpose is to derive the vacuum amplitude of electromagnetic field, which can be used to study the SE in any spherical confocal structure. To calculate the energy of spherical system we rewrite the fields as $E_s \rightarrow \frac{1}{2}(E_s + E_s^*) = \frac{1}{2}(A + A^*)F_s$ and $H_s \rightarrow \frac{1}{2}(H_s + H_s^*) = \frac{i}{2}(A - A^*)f_s$, where $s = r, \theta, \varphi$.

The fields energy in a spherical volume V is given by

$$W = \frac{1}{2} \int_{V} (\varepsilon_0 \varepsilon E^2 + \mu_0 H^2) dV = \beta_e^2 + \beta_h^2,$$
(6)

where β_e^2 and β_h^2 are the electric and magnetic parts of energy, which with help of (3) one can rewrite as

$$\beta_{e}^{2} = \frac{\varepsilon_{0}}{8} (A + A^{*})^{2} \int_{V} \varepsilon(r) (F_{r}^{2} + F_{\theta}^{2} + F_{\varphi}^{2}) dV, \\ \beta_{h}^{2} = -\frac{\varepsilon_{0}}{8} (A - A^{*})^{2} \int_{V} (f_{\theta}^{2} + f_{\varphi}^{2}) dV.$$
(7)

$$\beta_e^2 = \varepsilon_0 a_0^3 G_m^2 \{ I_m(\alpha) + \frac{1}{\varepsilon(\alpha)} R_m(\alpha) R_m'(\alpha) - \frac{1}{\varepsilon(0)} R_m(0) R_m'(0) \},$$
(8)

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$$\beta_h = \varepsilon_0 a_0^3 G_m^2 I_m(\alpha), \tag{9}$$

where $\alpha = k_0 a_0 = \omega a_0 / c$ and quantity

$$I_m(\alpha) = \frac{1}{\alpha^3} \int_0^\alpha R_m^2(y) dy,$$
(10)

is determined by the radial structure of field only. The angular part G_m^2 in (8)-(9) can be calculated in general form $G_m^2 = \int_0^{2\pi} d\varphi \int_0^{\pi} d\theta \sin \theta [\frac{1}{\sin^2 \theta} (\frac{\partial Y_m^j}{\partial \varphi})^2 + (\frac{\partial Y_m^j}{\partial \theta})^2] = m(m+1)$. Note for sphere with radius a_0 the boundary conditions $R_m(0) = 0$, $R'_m(0) = 0$ and $R_m(\alpha) = 0$, $R'_m(\alpha) = 0$ completely determines a spectrum of eigenfrequencies $\alpha_{vm} = \omega_{vm} a_0 / c$. A first condition corresponds to the fields limitation in the center r = 0, while the second reflects vanish the transverse fields at $r = a_0$. With Eqs. (8), (9) the latter means that in the boundary of sphere $r = a_0$ the Poynting vector flux is zero and the full fields energy in volume V is preserved (W = const). One can see from (8)-(9), when such conditions obey, the important rigorous equality of electric and magnetic energies follows $\beta_e^2 = \beta_h^2$. Note the form of equality $\beta_e^2 = \beta_h^2$ does not change for more complex multilayered structures. This becomes a start point to field's quantization in sphere. We write down such equality in the next form

$$\beta^{2} = \beta_{e}^{2} / a_{0}^{3} = \beta_{h}^{2} / a_{0}^{3} = m(m+1)I_{m}(\alpha).$$
(11)

Equation (11) generalizes the formula of equality of the energy of electric and magnetic fields of the plane-waves case [22] to a spherical geometry case. Now W in (6) can be rewritten as

$$W = a_0^3 \frac{\beta^2}{8} \varepsilon_0 \{ (A + A^*)^2 - (A - A^*)^2 \}.$$
 (12)

In treating (12) we find it convenient to introduce $A = \mathbf{E}a$, $A^* = \mathbf{E}a^+$, where **E** has unit of field, a^+ and a are creation and annihilations field operators ($[a,a^+]=1$) in the appropriate modes with frequency ω . After that W in (12) becomes the fields Hamiltonian operator with expectation value $2a^3\mathbf{E}^2\beta^2\frac{\varepsilon_0}{4}(n+1/2) = \hbar\omega(n+1/2)$. From the latter the amplitude $\mathbf{E} = \mathbf{E}(\alpha)$ for state $|0\rangle$ can be written in the next form

$$\mathbf{E}(\alpha) = \mathbf{E}_0 \Delta_m(\alpha), \ \Delta_m(\alpha) = \left(\frac{16\pi}{3\beta^2}\right)^{1/2} = \left(\frac{16\pi\alpha^3}{3m(m+1)I_m(\alpha)}\right)^{1/2}$$
(13)

where $\mathbf{E}_0 = \left(\frac{\hbar\omega}{2\varepsilon_0 V}\right)^{1/2}$ is well-known amplitude of the vacuum field (field per photon) for a plane geometry case [9].

Which does not dependent on the structure of field, and $V = 4\pi a_0^3/3$ is a volume of sphere. The quantity $\Delta_m(\alpha)$ in (13) defines the correction of such amplitude due to the spherical geometry. Equation (13) provides the solution of the vacuum field problem. One can see the amplitude $\mathbf{E}(\alpha)$ in the spherical geometry depends on the spherical *m* quantum number and also on the radial quantum number ν through the eigenfrequencies $\alpha_{\nu m}$. Formula (13) has general form and is valid for any confocal spherical structures deposited on the surface of microsphere. Now with the help of (13) one can write down the final expression for the tangential electric field in microsphere as following

$$E_{\theta} = \mathbf{E}_{0} \cdot \left(\frac{16\pi\alpha_{vm}^{3}}{3m(m+1)I_{m}(\alpha_{vm})}\right)^{1/2} \frac{1}{\alpha_{vm}^{2} z \varepsilon(z)} \frac{\partial R_{m}(\alpha_{vm} z)}{\partial z} \frac{\partial Y_{m}^{j}(\theta, \varphi)}{\partial \theta} (a_{vm}^{+} + a_{vm}), \quad (14)$$

where $z = r/a_0$, $\alpha_{vm} = \omega_{vm}a_0/c = k_{vm}a_0$, a_{vm}^+ and a_{vm} are creation and annihilations field operators in the modes with eigenfrequencies ω_{vm} of the corresponding spherical structure.

Formula for E_{φ} can be received from (3) by the change $\frac{\partial}{\partial \theta} \rightarrow \frac{\partial}{\sin \theta \partial \varphi}$. Expression (14) essentially differs from well-known plane-wave case [8], [9], [22]. It should be used since studying the SE in microspheres. Moreover, in complex spherical structures one can expect new features in view of essential modification of a photons spectrum due to the various interferential effects in spherical layers [23]. Generally, the evaluation $I_m(\alpha)$ in (10) requires the knowledge of the structure of the internal fields in multilayers. Various approaches for analysis of multilayered microspheres were proposed last time [23]-[25]. The detailed calculation one has to produce already beyond the continuum mode approximation. In [26] the analysis of SE was done by means of the direct numerical solution of the equations of the probability amplitudes of the atoms states.

3. Results and Discussion

Further concreteness in the definition of $I_m(\alpha)$ requires the knowledge of details the structure of electromagnetic oscillations. We apply the derived formulas to a simplest case of a hollow metallized microsphere with $\varepsilon = 1$, in this case $\mathbf{E}(\alpha)$ can be calculated in a closed form. For such a microsphere the solution of the radial field equation has form $R_m(y) = (\frac{2y}{\pi})^{1/2} J_{m+1/2}(y)$, where $J_m(y)$ is Bessel functions, $y = k_0 r$. Due to the perfectly conducting walls the boundary conditions $E_{\theta} = 0$ and $E_{\varphi} = 0$ at $r = a_0$ for *TE* waves result in the eigenfrequency equation $R_m(\alpha) = 0$ in form $J_{m+1/2}(\alpha) = 0$. The solution of this equation we write as α_{vm} , where v is a radial quantum number. In result $I_m(\alpha)$ in (10) can be reduced to $I_m(\alpha) = [J'_{m+1/2}(\alpha)]^2/2$. After that $\mathbf{E}(\alpha)$ in () for *TE* waves becomes $\mathbf{E}(\alpha) = \mathbf{E}_0 [m(m+1)\frac{\pi\alpha}{16} J_{m+1/2}^2(\alpha)]^{-1/2}$, $\alpha = \alpha_{vm}$.

Similar calculations for TM waves provide with the eigenfrequencies equation $J_{m+1/2}(\alpha) + 2\alpha J_{m+1/2}(\alpha) = 0$. In this case $\mathbf{E}(\alpha)$ in (13) has the next form $\mathbf{E}(\alpha) = \mathbf{E}_0 \left[m(m+1) \frac{\pi \alpha}{4} \left(1 - m \frac{m+1}{\alpha^2} \right) J_{m+1/2}(\alpha) \right]^{-1/2}, \ \alpha = \alpha_{vm} \text{ . If } \alpha_{vm} >> 1 \text{ from above follows,}$ that $\mathbf{E} \sim \mathbf{E}_0 / m$, i.e. the field per photon decreases with growth m quantum number. To write the asymptotic of the received formulas for a case $\alpha_{vm} >> m$ and $\alpha_{vm} z >> 1$ we use the asymptotic of Bessel functions [27] $J_{m+1/2}(\alpha) \sim (\frac{2}{\pi \alpha})^{1/2} \sin(\alpha - \frac{m\pi}{2})$. In this case (14) $E_{\theta} = \mathbf{E}_{0} \left[\frac{32\pi}{m(m+1)3} \right]^{1/2} \frac{\cos(\alpha_{vm}z - \frac{m\pi}{2})}{z} \frac{\partial Y_{m}^{j}(\theta, \varphi)}{\partial \theta} \left(a_{vm}^{+} + a_{vm} \right).$ From above formulas one can becomes derive the approximate values for eigenfrequencies: for easily TM case $\alpha_{vm} = (\pi/2)(m+2\nu-1)$, and for *TE* case $\alpha_{vm} = (\pi/2)(m+2\nu)$, where $m = 1, 2..., \nu = 1, 2...$. However the numerical solution of eigenfrequencies equations shows, that for large mthese formulas longer are not valid. Further, we the received formulas for the radial fields distribution analyzed numerically. In Fig.2 the radial distribution of the quantized tangential field $E_{\theta}(\alpha_{\nu m}r/a_0)/\mathbf{E}_0$ in (14) for various *m* is shown at corresponding values of eigenfrequencies α_{vm} , calculated numerically.



Fig. 2. Radial distribution of the tangential field $E_{\theta}(\alpha_{vm}r/a_0)/E_0$ for TM case in the metallized microsphere for various radial v=1..10 and spherical m quantum numbers. a) m=1; b) m=10; c) m=30; c) m=100; α_{vm} are the dimensionless eigenfrequencies (see text), a_0 is the radius of microsphere.

One can see from Fig.2, only at m = 1 the first maximum of the field is placed in the center of microsphere. For large m the maximums migrate to periphery, and the field in a vicinity of the center is exponentially small.

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For large m = 100 practically all fields are concentrated on the periphery (see Fig.2,d). Due to strong oscillations the fields nodes at large m >> 1 become quite intricate and the rate of SE can be very sensitive to the localization of an atom in microsphere.

4 Conclusion

In conclusion the quantization electromagnetic field in microsphere is made with taking into account the spatial structure of field explicitly. The exact expression for vacuum fields amplitude, associated with the spherical geometry is derived. It is found, that the vacuum field amplitude strongly depends on the fields' spherical quantum number m and since m >> 1 the field varies as 1/m. This work is supported by Mexican National Council of Science and Technology (CONACyT project #35455-A).

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