

Research Article

The Subwavelength Optical Field Confinement in a Multilayered Microsphere with Quasiperiodic Spherical Stack

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We study the frequency spectrum of nanoemitters placed in a microsphere with a quasiperiodic subwavelength spherical stack. The spectral evolution of transmittancy at the change of thickness of two-layer blocks, constructed following the Fibonacci sequence, is investigated. When the number of layers (Fibonacci order) increases, the structure of spectrum acquires a fractal form. Our calculations show the radiation confinement and gigantic field enhancement, when the ratio of layers' widths in twolayer blocks of the stack is close to the golden mean value.

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1. Introduction

The use of microcavities and microspheres in advanced optoelectronics has provided a new view of various effects and interactions in highly integrated, functional photonic devices. A main fundamental question in this area is how to drastically increase the spectral optical field strength, using artificially produced alternating layers on a surface of microsphere. Nowadays the basic regime of the operation for bare (uncoated) dielectric microspheres is the whispering gallery mode (WGM). The extremely high-quality factors (Q -factors) $\sim 10^8 \div 10^9$ have been realized [1–3]. But since fabricating the coated dielectric spheres of the submicron sizes, the problem arises to study the optical oscillations in microspheres beyond the WGM regime for harmonics with small spherical numbers.

It is well known that a bare dielectric sphere has a complex spectrum of the electromagnetic low-quality (Q factor) eigenoscillations because of the energy leakage into the outer space [4]. The case of the compound structure, when the dielectric sphere is coated by an alternative stack, is much richer. The Q -factor of optical oscillations has a large value in the frequency regions of weak transmittancy, and beyond these regions Q remains small [5–8]. This gives rise to a large variety of optical properties of microspheres, coated with a multilayer stack. Such a system can serve as

a spherical symmetric photonic band gap structure, which possesses strong selective transmittance properties [9, 10], with incorporating the nanometer-sized photon emitters. These possibilities allow to expand essentially the operational properties of microspheres with attractive artificial light sources for advanced optical technologies. It is now feasible to construct such a microsphere accurately, and the parameters may be precisely controlled and measured (see [11] and reference therein). Equally important, this system can provide a compact and simple building block for studying the quantum aspects of light. The attachment of semiconductor nanoclusters onto a spherical microcavity already has allowed the observation of the vacuum Rabi splitting [12].

Among the various quasicrystals, the Fibonacci 1D and Penrose 2D structures have been the subject of an extensive theoretical and experimental efforts [13–19]. However, though the plane Fibonacci layers are studied well enough, the properties of quasiperiodic spherical structures were not studied sufficiently yet.

The important optical property of a periodic alternating spherical stack is a possibility to confine the optical radiation. However, is periodicity necessary for such resonant optical effects?

In order to answer this question, we have studied the optical radiation of a nanosource (nanometer-sized light

source), placed into a microsphere coated by a quasiperiodic multilayered structure (stack) constructed following the Fibonacci sequence. In such a system the 1D (radial depending) theory is strictly valid. Such structures are called quasiperiodic, and are lying outside the constraints of periodicity. One of the main properties of such a stack is reflection of the electromagnetic waves from the interfaces of the layers, that results in the collective wave contributions. The collective optical effects in a quasiperiodic spherical stacks are appreciated only if number of layers in the stack is large enough. In this case, various approximations based on the decomposition of field states in the partial spherical modes have insufficient accuracy, so the deeper insight requires more advanced approaches. Our approach is based on dyadic Green's function technique [20], that provides an advanced approximation for a multilayered microsphere with nanoemitters [21]. We have applied this approach to a quasiperiodic spherical stack and found the substantially enhanced optical resonances (Green function strength), when the ratio of layers' widths in two-layer blocks in stack (quasiperiodicity parameter) is close to the golden mean value. As far as the authors are aware, the optical fields of nanoemitters placed in a microsphere with quasiperiodic spherical stack still have poorly been considered, though it is a logical extension of previous works in this area.

This paper is organized as follows. In Section 2, we formulate our approach and basic equations for optical fields in a dielectric multilayered microsphere. We outline the numerical scheme of applying the dyadic Green function (DGF) technique to evaluate the spectrum of a nanoemitter placed in such a quasiperiodic system. In Section 3, we present our numerical results on structure of the cavity field states and resonances in a microsphere with quasiperiodic spherical stack dependently of the quasiperiodicity parameter. We found the enhanced field peaks if such a parameter is close to the golden mean value. In the last section, we summarize our results.

2. Basic Equations

A 1D quasiperiodic (QP) spherical stack, where a Fibonacci sequence is considered, can be constructed following a simple procedure. Let us consider two neighbor 2-layer segments, long and short, denoted, respectively, by L and S . If we place them one by one onto surface of a microsphere, we obtain a sequence: LS . In order to obtain a QP sequence, these elements are transformed according to Fibonacci rules as follows: L is replaced by LS , S is replaced by L : $L \rightarrow LS$, $S \rightarrow L$. As a result, we obtain a new sequence: LSL . Iteratively applying this rule, we obtain, in the next iteration, a sequence with a five-element stack $LSLLS$, and so on. One can control the properties of such a QP stack by the use of some control parameter (see factor γ later in Section 3). For the stack with N -elements, where $N \gg 1$, the ratio of numbers of long to short elements is the golden mean value, $\Gamma_0 = (1 + \sqrt{5})/2 \approx 1.618$.

Generally in spherical geometry the wave field depends on position of a source and it is formed on a distance scale

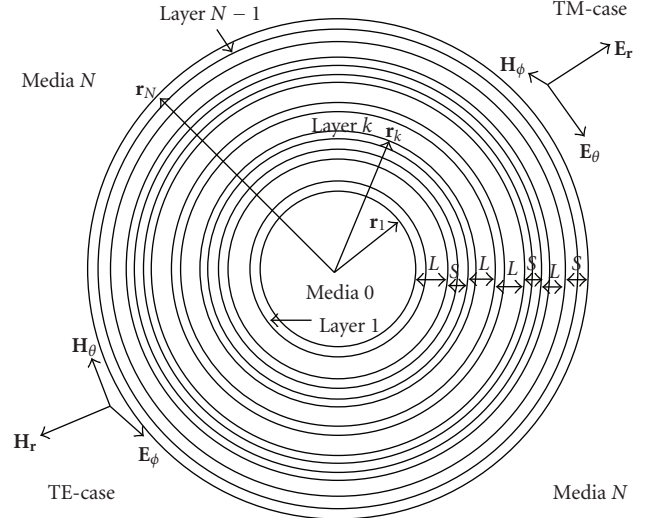


FIGURE 1: Geometry of multilayered microsphere. Stack of quasiperiodic multilayers is deposited on the surface of microsphere.

of the order magnitude of the radius of microsphere or thickness of nanolayers of a stack. For analysis of such a spectrum it is necessary to use more advanced approach: the Green function method. The base of the latter is representation of optical field, radiated by a nanosource in a coated microsphere, as a weighed superposition (sum) of forward and backward waves (reflected from the layers interfaces). We consider a situation when radiating point source (nanoemitter) is placed into microsphere coated by a quasiperiodic stack. In this case, the frequency spectrum is not described longer by a spectrum of bare microsphere slightly perturbed by the external stack. If the number of layers is large enough, we have to study the photons field taking into account the spectral contributions both bottom microsphere and a quasiperiodic stack. In order to calculate the properties of such a field, we apply the Green function technique. In this case, a nanosource corresponds to a nanorod or quantum dot that recently were employed in experiments with microspheres (see [12, 22] and references therein).

The spatial scale of the nanoemitter objects ($\sim 1-100$ nm) is in at least of one order of magnitude smaller than the spatial scale of microspheres ($\sim 10^3-10^4$ nm). Therefore, in the coated microsphere (Figure 1), we can represent the nanoemitter structure as a point source placed at \mathbf{r}' and having a dipole moment \mathbf{d}_0 . It is well known that the solution of the wave equation for the radiated electromagnetic field \mathbf{E} due to a general source $\mathbf{J}(\mathbf{r}')$ reads ($\mu = 1$) [23, 24]

$$\mathbf{E}(\mathbf{r}) = i\omega\mu_0 \int_V d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \cdot \mathbf{J}(\mathbf{r}'), \quad (1)$$

where $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ is the dyadic Green function (DGF), which depends on the type of the boundary conditions imposed on $\mathbf{E}(\mathbf{r})$ and contains all physical information necessary for describing the multilayered structure (the time dependence is assumed to be as $e^{i\omega t}$). Equation (1) is complemented by

the standard boundary conditions: limitation of fields in the center of microsphere, continuity of tangential components of fields at the interfaces of layers. Also, we use Sommerfeld's radiation conditions, there is only outgoing wave in the external boundary of a microsphere. In this case, the electromagnetic field \mathbf{E} in the coated structure consists of sum of the waves radiating in the surrounding medium and the multiple wave reflections due to interfaces between layers. Substituting the nanorod source in the form $\mathbf{J} = i\omega\mathbf{d}$, $\mathbf{d} = \mathbf{d}_0\delta(\mathbf{r} - \mathbf{r}')$ in (1), we obtain

$$\mathbf{E}(\mathbf{r}, \mathbf{r}', \omega) = -\mathbf{p}_0\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega), \quad (2)$$

where $\mathbf{p}_0 = (\mathbf{d}_0/\varepsilon_0)(\omega^2/c^2)$, \mathbf{r} is the point where the field is observed, while \mathbf{r}' is the nanorod (point nanoemitter) location. In such a situation, the nanoemitter frequency spectrum is identical to dyadic Green's function (DGF) spectrum.

Let us consider the multilayered spherical structure: a concentric system of spherical layers contacting with the sphere (a concentric stack) deposited onto the surface of the microsphere with a nanoemitter placed in such a structure (see Figure 1). The layers are localized at the distances R_k from the center, where $d_k = R_{k+1} - R_k$ is the width of a k th layer.

Let us first specify some details of the Green function technique for multilayered microspheres and introduce our notations. Following the approach [20], we write down DGF of such a system as follows:

$$\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{G}^V(\mathbf{r}, \mathbf{r}', \omega)\delta_{f_s} + \mathbf{G}^{(f_s)}(\mathbf{r}, \mathbf{r}', \omega), \quad (3)$$

where $\mathbf{G}^V(\mathbf{r}, \mathbf{r}', \omega)$ represents the contribution of the direct waves from the radiation sources in the unbounded medium, whereas $\mathbf{G}^{(f_s)}(\mathbf{r}, \mathbf{r}', \omega)$ describes the contribution of the multiple reflection and transmission waves due to the layer interfaces. The dyadic Green tensor $\mathbf{G}^V(\mathbf{r}, \mathbf{r}', \omega)$ in (3) is given by

$$\begin{aligned} \mathbf{G}^{(f^e)}(\mathbf{r}, \mathbf{r}', \omega) \\ = \frac{ik_s}{4\pi} \sum_{p=e,o} \sum_{m=1}^{\infty} \sum_{l=0}^m \frac{2m+1}{m(m+1)} \frac{(m-l)!}{(m+l)!} (2 - \delta_{0l}) \mathbf{G}_{pml}^{(f,e)}(\mathbf{r}, \mathbf{r}', \omega), \end{aligned} \quad (4)$$

where $\mathbf{G}_{cml}^{(f,e)}(\mathbf{r}, \mathbf{r}', \omega)$ is the particular Green tensor [20, 21], m is the spherical and l is the azimuth quantum numbers, $k_i = \omega n_i/c$, $n_i = \sqrt{\varepsilon_i(\omega)}$ is a refraction index. It is worth to note that in a spherical case, the configuration of photon field strongly depends on the position of source in a coated microsphere. Such a field has a structure of single spherical wave only if a nanosource is placed very close to the center [6]. Otherwise, many forward and backward (reflected) spherical waves contribute, therefore the complete description of such a system or the Green function (3), (4) has quite complicated structure.

As already we have mentioned, we represent a nanorod by a dipole nanoemitter. Since nanorods are highly polarized objects, we pay more attention to the case when the dipole orientation of a nanoemitter is $\mathbf{d} = d\hat{\boldsymbol{\varphi}}$ [22], so only the

tangential components of the Green tensor $G_{\varphi\varphi}$ contribute. For a spherical stack, $G_{\varphi\varphi}(\mathbf{r}, \mathbf{r}, \omega)$ is expressed through the complex Hankel functions [20, 23]; therefore an analytical study of the spectrum is rather difficult problem. Further we use numerical approach; the numerical scheme for our study is similar to those used in [21].

3. Numerical Results

In this section, we study the frequency spectrum of a nanoemitter radiation for quasiperiodic Fibonacci layers (spherical stack) deposited on the surface of a microsphere (Figure 1) for different numbers of layers in the stack. First, let us remind that the Green function relates to energy of a fluctuating electromagnetic field strength $\mathbf{E}(\mathbf{r})$ (at small dissipation) as [25] $\langle \mathbf{E}(\mathbf{r})^2 \rangle = (\hbar\omega^2/c^2) \coth(\hbar\omega/2T_0) \text{Im}(G(\mathbf{r}, \mathbf{r}, \omega)) \rightarrow_{T_0 \rightarrow 0} (\hbar\omega^2/c^2) \text{Im}(G(\mathbf{r}, \mathbf{r}, \omega))$, where $G(\mathbf{r}, \mathbf{r}, \omega)$ is the Green function, that is, $G_{\varphi\varphi}(\mathbf{r}, \mathbf{r}, \omega)$ in our case, T_0 is temperature. We note that such a field state \mathbf{E} is not a photonic state in general, but a state of the macroscopic medium, dressed by the electromagnetic field [25, 26].

The following parameters have been used in our calculations: the geometry of system is $A\{L(B, C) \cdots S(B, C) \cdots\}D$, where letters A, B, C, D indicate the materials in the spherical stack, respectively. The bottom microsphere has refraction index $n_A = 1.5 + 2 \cdot 10^{-4}i$ (A , glass, radius 1000 nm). The distinct two-layered blocks ($L(B, C)$ and $S(B, C)$) are stacked according to the Fibonacci generation rule. For L and S blocks, we use the notation $L = (B, C, 1)$ and $S = (B, C, \gamma)$, where γ is the ratio of both thicknesses: $\gamma = (C)_S/(C)_L \leq 1$. Refraction indices of the layers in blocks are $n_B = 3.58 + 9 \cdot 10^{-4}i$ (Si, width 122 nm), $n_C = 1.46 + 10^{-3}i$ (SiO₂, width 300 nm) [27], and $n_D = 1$ (D , surrounding space). For L -block layers B and C are constructed as $\lambda/4$ layers, while for S -block thickness B is the same as for L -block, but the thickness C is 150 nm ($\gamma = 0.5$). For example, for Fibonacci order F_9 , the total thickness of the microsphere with 68 layers is 13.4 μm (34 two-layer blocks). To consider the realistic layer case, we have added to each n_i a small imaginary part that corresponds to a material dissipation.

In order to study the behavior of field in the microsphere, we have calculated the frequency spectrum of the transmittance coefficient T and corresponding spectrum of imaginary parts of the Green function $W \equiv \text{Im}(G_{\varphi\varphi}(\mathbf{r}, \mathbf{r}, \omega))$, where \mathbf{r} is position of a nanoemitter, $\omega = 2\pi f$. We have calculated evolution of a spectrum for different values γ , and also for different number of layers in the spherical stack (Fibonacci order F_n) for a range [300–600] THz or [1000–500] nm (DGF is normalized on radius of the bottom microsphere). Most intensive optical peak was found for F_9 stack, when $\gamma = 0.618 = 1/\Gamma_0$ ($\Gamma_0 \approx 1.618$ is golden mean value). The details of field spectra are shown in Figure 2 for 68 layers in the stack (34 of 2-layer blocks, order Fibonacci is F_9). We observe from Figure 2(a), that such a spectrum consists of peaks with various amplitudes, however the most intensive peak with $\text{Im}(G_{\varphi\varphi}) \approx 87$ is located at 436.1 THz (details of this peak are shown in the inset). We also have calculated that such a peak relates to eigenfrequency of the system $f = 436.098$ THz with quite large-quality factor

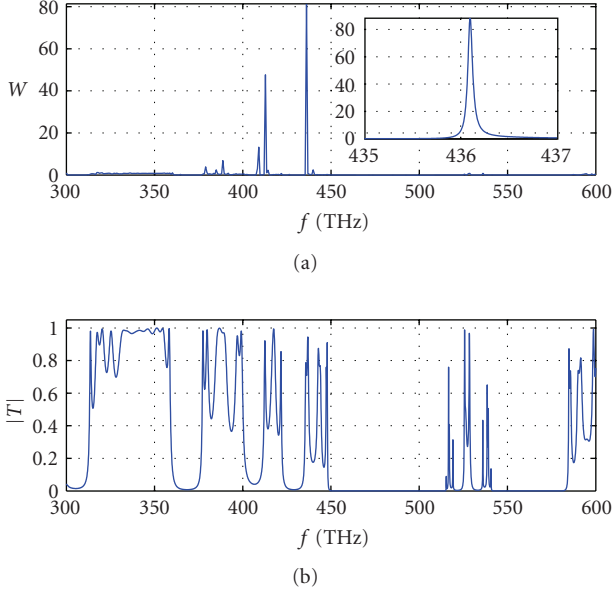


FIGURE 2: (a) Imaginary parts of tangential component of the Green function $W = G_{\varphi\varphi}(r, r)$ for $r = 900$ nm, (b) Frequency spectrum of transmittance coefficient T for spherical quantum number $m = 9$. Microsphere is coated by quasiperiodic stack with $N = 68$ (34 of 2-layers blocks, order Fibonacci F_9) for $\gamma = 0.618$. See details in text.

$Q = 7369.02$. Thus, even though periodicity of the stack is broken, well-defined intensive peak of field is clearly seen. Small peaks in Figure 2(a) have rather indented form due to the contribution of several close small resonances corresponding to different spherical modes for DGF in (3)-(4). The transmittancy spectrum for $\gamma = 0.618$ is shown in Figure 2(b). We observe that as opposed to just a periodic $\lambda/4$ case [6, 7, 28], the Fibonacci stack has a fractal-like transmittancy spectrum, see Figure 2(b). Such a spectrum consists of narrow resonances separated by numerous pseudoband gaps, induced by incoherent re-reflections of optical waves from quasiperiodic layers interfaces. Resonances correspond to poles of DGF at complex eigenfrequencies f , where $\text{Im}(f)$ determinates the width of a resonance or Q -factor of oscillations $Q = \text{Re}(f)/2\text{Im}(f)$. At small dissipation, peaks have a typical Lorentzian line shape $\text{Im}(G_{\varphi\varphi}) \sim \varepsilon/(\delta^2 + \varepsilon^2)$, where δ is a detuning from a resonance, and ε is the linewidth.

Adding more layers should strongly narrow the resonances. The spectrum T becomes more complicated and new resonances do form a well-expressed fractal of transmittancy. Corresponding spectrum of the Green function becomes richer, since a greater number of eigenmodes contribute into numerous pseudoband gaps of T . It is important to note that, as one can see from Figure 2(b), the cavity modes excited by a dipole inside a coated microsphere are distinctly different from whispering gallery modes characterized by $m \gg 1$ and $T \rightarrow 0$.

We have calculated evolution of a spectrum for different values γ close to $\gamma = 1/\Gamma_0$ for fixed number of layers in the spherical stack (Fibonacci order F_n). The results are

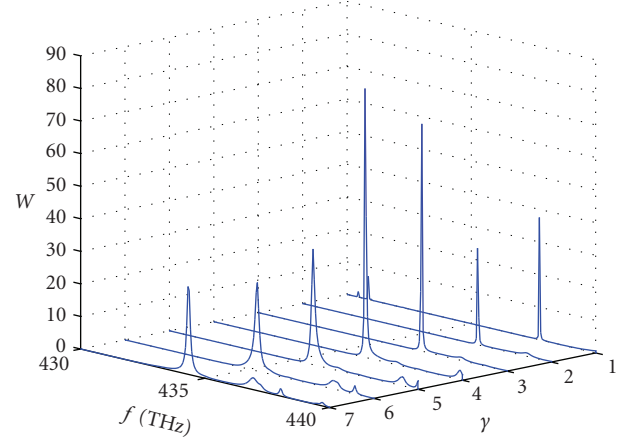


FIGURE 3: Spectrum of imaginary part of the Green function $W = G_{\varphi\varphi}(r, r)$ for $r = 900$ nm and 68 layers in stack (34 of 2-layer blocks F_9) for different values of parameter quasiperiodicity γ close to the golden mean value: (1) 0.55, (2) 0.58, (3) 0.6, (4) 0.618, (5) 0.63, (6) 0.65, and (7) 0.68.

displayed in Figure 3 for a range [430–440] THz, or [0.70–0.68] μm . One can see that in this area the spectrum consists of rather narrow resonances, and a set of satellite peaks appears around the main peak. The intensity of this peak changes for different γ , giving rise to another main peak. When γ approach to $1/\Gamma_0 = 0.618$, the field peaks become sharper, as shown in Figure 3. We observe that the amplitude of resonances is maximal for case $\gamma = 0.618$.

It is of great interest to compare the spectra in Figure 2 to the case with distinct “quasiperiodic parameter” γ , especially to the case $\gamma = 1$ case, when QP stack effectively becomes a periodic $\lambda/4$ spherical stack. In result of calculations, we have found that the dominating peak for $\gamma = 0.618$ (Figure 2(a)) is already 40 times higher with respect to periodic $\lambda/4$ case, that is, a signature of a quasiperiodicity strength in a coated microsphere cases.

In previous figures, the frequency spectrum of the field ($\sim \text{Im}(G_{\varphi\varphi}(r, a, f))$) for the quasiperiodic stack was shown. The fractal complicity of the transmittancy spectrum is defined by the intrinsic properties of the quasiperiodic spherical stack independently on the nanoemitter location. However in experiments, it is important to identify the spatial distribution of the field, radiated by nanosources located in such a quasiperiodic microsphere. Therefore, it is of interest to consider the spatial field distribution in a cross-section ($r, \varphi, \theta = \text{const}$) that contains both center of the coated microsphere and nanoemitter for some resonant frequency. Such a distribution is shown in Figure 4 for $\theta = 1$ and the most intensive resonance at $f_0 = 436.098$ THz (see Figure 2(a)), when the quasiperiodicity parameter $\gamma = 0.618$. We observe from Figure 4 that $W(r, \varphi) = \text{Im}(G_{\varphi\varphi}(r, a, \varphi))$ has a very sharp peak in the place of the nanosource location. Such a spatial field structure may be treated as a confinement of the electromagnetic energy $\sim \text{Im}(G_{\varphi\varphi}(\mathbf{r}))$ inside the coated microsphere. The leakage of photons through such a structure into the outer space obviously is

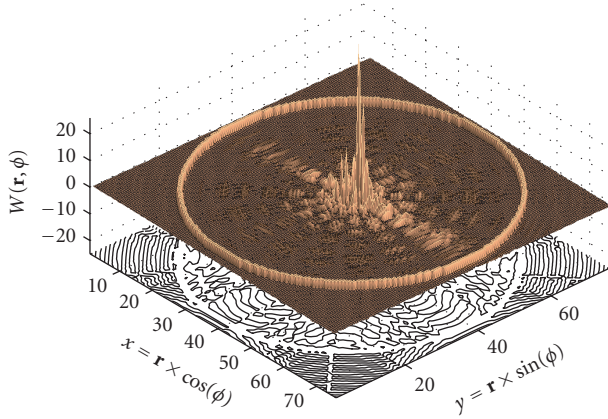


FIGURE 4: Spatial structure $W(r, \varphi) = \text{Im}(G_{\varphi\varphi}(r, a, \varphi))$ in a cross-section $0 < r < 21 \mu\text{m}$ and $0 < \varphi < 2\pi$ of the microsphere with quasiperiodic stack for eigenfrequency $f = 436.09 \text{ THz}$. A nanoemitter is placed at point $a = 900 \text{ nm}$. Other parameters are same as in Figure 2. One can observe the confinement of field in the stack. Outer cycle only indicates the external boundary $R_{\text{ext}} = 13.8 \mu\text{m}$ of the quasiperiodic spherical stack.

small. We observe from Figure 4 that the field structure inside of quasiperiodic stack is anisotropic and quite intricate, but the field distribution beyond the coated microsphere has a periodic character.

4. Conclusion

We have studied the frequency spectrum of nanoemitters placed in a microsphere with a quasiperiodic subwavelength spherical stack. We found that the transmittancy spectrum of such a stack consists of quasi-band gaps and narrow resonances, induced by re-reflection of optical waves. The spectral evolution of transmittancy at the change of the thickness of two-layer blocks, constructed following the Fibonacci sequence, is investigated. When the number of layers (Fibonacci order) increases, the structure of spectrum acquires a fractal form. We show that the width of resonant peaks in the frequency spectrum becomes *extremely narrow* for a quasiperiodic spherical stack of a high Fibonacci order. In principle, that allows creating a narrow-band filter with a transmission state within the forbidden band gap of nanoemitters, incorporated in such a coated microsphere. We have found the confinement and the gigantic enhancement of the optical field in quasiperiodic structure, when the ratio of layers' widths in two-layer blocks of stack is close to the golden mean value. This allows to confine resonantly the field energy in the quasiperiodic stack in very narrow frequency range in order to create very selective stop-band filters. Incorporating nanoemitters into such structured microspheres can open new opportunities for the active control of light nanosources.

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