



REGULAR ARTICLE

The Purcell Factor of a Dipole Molecule Located Near a Spherical Metal Nanoparticle

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The paper considers the problem of the effect of a spherical metal nanoparticle on the spontaneous radiation of a dipole molecule located in the vicinity of the nanoparticle. Expressions for the frequency and size (at the surface plasmon resonance frequency) dependences of the Purcell factor, as well as the coupling factor with the resonator, were obtained. It is the nanoparticle that acts as a resonator. The Drude model is considered suitable for the frequency dependence of the dielectric function. Calculations were performed for spherical nanoparticles of different radii and made of different metals, located in different dielectric media and at different distances from the dipole molecule, taking into account volume and surface relaxation and radiation attenuation.

According to the results of the calculations, it was established that the frequency dependences of the Purcell factor and the coupling factor with the resonator are characterized by two maxima. One of them, with a relatively small width, is observed at a frequency that practically coincides with the frequency of the surface plasmon resonance of a single spherical metal nanoparticle. The second, broad, is in the red or infrared region of the spectrum. It is proved that the reason for the emergence of this second low-frequency maximum is the resonance of the "whispering gallery" modes. At the same frequency, the greater the radius of the nanoparticle and the smaller the distance from it to the dipole molecule, the larger the investigated factors are. It has been demonstrated that a change in the material of a nanoparticle results in a change in the value of the Purcell factor and a shift of the maxima in its frequency dependence. The reason for this effect is a significant difference in the plasma frequency and the fraction of interband transitions in the dielectric function for different metals. The effect of the environment in which the silver nanoparticle-dipole system is located on the frequency and size dependence of the Purcell factor was studied. It was established that with an increase in the dielectric constant of the medium, the value of the Purcell factor decreases and the maxima of its frequency dependence shift to lower frequencies.

**Keywords:** Purcell factor, Dipole molecule, Metal nanoparticle, Surface plasmon resonance.

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1. INTRODUCTION

In connection with the rapid development of nanotechnology, the question of the influence of nanoparticles on optical phenomena in their vicinity arose. Small sizes result in the concentration of electromagnetic fields near the nanoparticles. Therefore, nanoparticles can be effectively used to influence the fluorescence and spontaneous emission of nearby atoms (molecules).

In practice, problems about the effect of nanoparticles on spontaneous radiation arise when studying individual molecules using aperture and apertureless scanning microscopes [1, 2]. Nanoparticles are also used to control fluorescence [3-6].

It is known that when the size of a nanoparticle is lower or comparable to the wavelength, the rate of spontaneous radiation of a molecule located near the nanoparticle can be increased and decreased compared to the rate of spontaneous radiation in vacuum. This is the essence of the Purcell effect [7, 8]. Quantitatively, this effect is characterized by the Purcell factor, which is defined as the ratio of the rate of spontaneous radia-

tion changed by the environment to the rate of spontaneous radiation in vacuum.

The Purcell effect affects physical processes in nanolasers (spasers), manifesting itself in the spatial redistribution of spontaneous radiation: suppression of radiation in certain modes and its amplification in others. In the presence of feedback in the system, there is an amplification of spontaneous radiation proportional to the Q factor of the resonator. At the same time, the Purcell factor can have a significant value [9]. For 1D and 2D systems, provided that the Q factor of the resonator is limited only by internal losses in the metal, the Purcell factor reaches the value  $10^2$ . It was shown [10-12] that in plasmonic nanostructures the Purcell factor varies within  $10 - 1 \cdot 10^3$ .

The Purcell factor is significant at frequencies close to the frequencies of surface plasmon resonance (SPR), at which the maximum localization of the field is realized, but the losses are also maximum. For 0D systems (nanoparticle-antenna), the value of the Purcell factor can reach  $10^6$  with limitations arising from non-local

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effects [9, 13-15].

The interaction of light with a quantum emitter (molecule) can be increased by reducing the volume of the optical mode and increasing the Q factor of the resonator [7]. In plasmon lasers, amplification can be achieved even with a low Q-factor of the resonator due to a small mode volume. In metal nanostructures, the effect of increasing the radiation speed is realized, as a rule, only for a few modes, mainly for the most localized ones. For a plasmon nanolaser, this means that the strongest coupling of the emitter is realized with the most localized modes of the nanolaser resonator.

The only known configuration of a plasmon nanolaser, in which all dimensions can be significantly smaller than the wavelength, is based on using a metal nanoparticle as a resonator. The size of the nanolaser in this case is determined only by the size of the particle-resonator and the gain factor of the active medium. A nanolaser based on a nanoparticle is implemented using localized surface plasmon resonance. In work [16], the excitation of such a resonance was observed in a nanoparticle surrounded by an active medium with a total size of 44 nm.

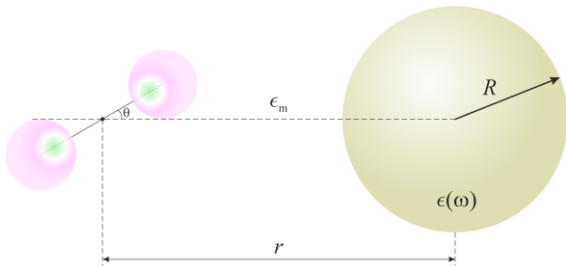
Small relaxation times in metals, small size of the resonator, short lifetime of surface plasmons, and a large Purcell factor ensure the achievement of generation frequency in the terahertz range [9, 17]. This makes plasmonic nanolasers attractive for many applications.

Of special interest is the question of using spherical metal nanoparticles as nanolaser resonators. In our previous works, the effect of oxide, passive and active dielectrics layers on the optical characteristics of spherical metal nanoparticles was analyzed [18-20]. The size and frequency dependences of the Q factor of metal particles of various geometries, including spherical one, were also studied [21, 22].

Thus, the problem of the effect of a metal nanoparticle on the spontaneous radiation of a molecule is prepared and it is proposed for consideration in this paper.

## 2. BASIC RELATIONSHIPS

Consider a system that consists of a spherical metallic nanoparticle of radius  $R$  and a dipole molecule located at a distance  $r$  from the center of the nanoparticle. We consider that the particle and the molecule are in a medium with dielectric permeability  $\epsilon_m$  (Fig. 1).



**Fig. 1** – Geometry of the problem

The quantities describing the optical properties of a nanoparticle-resonator are the Purcell factor  $\mathcal{F}_p$  and the coupling factor  $\beta$  of spontaneous radiation with the resonator, which are determined by the relations:

$$\mathcal{F}_p = \frac{8\pi^4 c^3 \omega_{sp}}{Q V_{sp} \omega^2 \left[ (\omega - \omega_{sp})^2 + (\omega_{sp}/Q)^2 \right]} \times \left( 1 + \frac{1 + 3 \cos^2 \theta \left( \frac{R}{r} \right)^6}{\epsilon_m^2} |\tilde{\alpha}|^2 \right), \quad (1)$$

$$\beta = \frac{\mathcal{F}_p}{1 + \mathcal{F}_p}, \quad (2)$$

where  $c$  is the velocity of light;  $\theta$  is the angle at which the dipole is oriented relative to the line connecting it to the center of the spherical particle; surface plasmon resonance frequency is

$$\omega_{sp} = \frac{\omega_p}{\sqrt{\epsilon^\infty + 2\epsilon_m}}; \quad (3)$$

the dimensionless polarizability of the nanoparticle is

$$\tilde{\alpha}(\omega) = \frac{\epsilon(\omega) - \epsilon_m}{\epsilon(\omega) + 2\epsilon_m}; \quad (4)$$

plasmonic mode volume is

$$V_{sp} = \left( \frac{\pi c}{\omega_{sp} \sqrt{\epsilon_m}} \right)^3; \quad (5)$$

Q factor of the nanoparticle-resonator is

$$Q = \frac{\omega}{2 \text{Im} \epsilon(\omega)} \frac{d \text{Re} \epsilon(\omega)}{d \omega}. \quad (6)$$

In the Drude model, the dielectric function of metal of the particle has the form

$$\epsilon(\omega) = \epsilon^\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma_{\text{eff}})}, \quad (7)$$

where  $\omega_p$  is the plasma frequency;  $\epsilon^\infty$  is the contribution of interband transitions. The effective rate of relaxation  $\gamma_{\text{eff}}$  is determined by the additive contributions of rates of bulk and surface relaxations and radiation damping [22]:

$$\gamma_{\text{eff}} = \gamma_{\text{bulk}} + \gamma_s + \gamma_{\text{rad}}, \quad (8)$$

where  $\gamma_{\text{bulk}} = \text{const}$ ,

$$\gamma_s = \mathcal{A}(\omega, R) \frac{v_F}{R}, \quad (9)$$

$$\gamma_{\text{rad}} = \frac{2}{9} \frac{\epsilon^\infty + 2\epsilon_m}{\sqrt{\epsilon_m(\epsilon^\infty + 2\epsilon_m)}} \left( \frac{\omega_p R}{c} \right)^3 \mathcal{A}(\omega, R) \frac{v_F}{R}. \quad (10)$$

In formulas (9) and (10),  $v_F$  is the Fermi velocity of electrons;

$$\mathcal{N}(\omega, R) = \frac{3}{4} \frac{1}{\epsilon^\infty + 2\epsilon_m} \left( \frac{\omega_p}{\omega} \right)^2 \times \left[ 1 - \frac{2v_s}{\omega} \sin \frac{\omega}{v_s} + 2 \left( \frac{v_s}{\omega} \right)^2 \left( 1 - \cos \frac{2v_s}{\omega} \right) \right] \quad (11)$$

is the coherence loss parameter, where  $v_s = v_F/2R$  is the frequency of individual electron oscillations.

Neglecting the oscillating terms in the square brackets of expression (11), relation (8) can be represented in the form

$$\gamma_{\text{eff}} = \gamma_{\text{bulk}} + \frac{\mathcal{N}}{\omega^2}, \quad (12)$$

where

$$\mathcal{N} = \frac{1}{2} \omega_p^2 \frac{v_F}{R} \left[ \frac{1}{2} + \frac{4}{9} \frac{1}{\sqrt{\epsilon_m(\epsilon^\infty + 2\epsilon_m)}} \left( \frac{\omega_p R}{c} \right)^3 \right]. \quad (13)$$

Let's find the size dependence of the Purcell factor at the surface plasmon resonance frequency ( $\omega = \omega_{sp}$ ). From expression (7) we get

$$\text{Re} \epsilon(\omega_{sp}) = -2\epsilon_m, \quad \text{Im} \epsilon(\omega_{sp}) \cong \frac{\omega_p^2 \gamma_{\text{eff}}^{sp}}{\omega_{sp}^3}. \quad (14)$$

The expressions for the Q factor and  $|\tilde{\alpha}|^2$  take the form

$$Q \cong \frac{\omega_{sp}}{\gamma_{\text{eff}}^{sp}}, \quad (15)$$

$$|\tilde{\alpha}|^2 \cong 1 + \left( \frac{3\epsilon_m \omega_{sp}^3}{\omega_p^2 \gamma_{\text{eff}}^{sp}} \right)^2, \quad (16)$$

where

$$\gamma_{\text{eff}}^{sp} = \gamma_{\text{bulk}} + \frac{\mathcal{N}}{\omega_{sp}^2}. \quad (17)$$

Substituting (15), (16) into (1), we get

$$F_p = 8\pi\epsilon_m^{3/2} \frac{\omega_{sp}}{\gamma_{\text{eff}}^{sp}} \left\{ 1 + \frac{1 + 3\cos^2 \theta}{\epsilon_m} \left( \frac{R}{r} \right)^6 \left[ 1 + \left( \frac{3\epsilon_m \omega_{sp}^3}{\omega_p^2 \gamma_{\text{eff}}^{sp}} \right)^2 \right] \right\}. \quad (18)$$

This relationship together with formulas (1) and (2) is the basis for obtaining numerical results.

### 3. RESULTS OF CALCULATIONS AND THEIR DISCUSSION

Calculations of the Purcell factor and the coupling factor with a particle-resonator were carried out for spherical nanoparticles of different radii made of dif-

ferent metals, different environments, and different distances between the nanoparticle and a dipole molecule. The parameters required for calculations are given in tables 1 and 2.

**Table 1** – The parameters of metals ( $a_0$  is the Bohr radius) (see, for example, [22, 23] and references therein)

Metals	Value				
	$r_s / a_0$	$m^* / m_e$	$\epsilon^\infty$	$\hbar\omega_p$ , eV	$\hbar\gamma_{\text{bulk}}$ , eV
Cu	2.11	1.49	12.03	12.6	0.024
Au	3.01	0.99	9.84	9.07	0.023
Ag	3.02	0.96	3.7	9.17	0.016
Pt	3.27	0.54	4.42	15.2	0.069
Pd	4.00	0.37	2.52	9.7	0.091

**Table 2** – Dielectric permittivity of matrices [22].

Substance	Air	CaF <sub>2</sub>	Teflon	Al <sub>2</sub> O <sub>3</sub>	C <sub>60</sub>
$\epsilon_m$	1.0	1.54	2.3	3.13	6.0

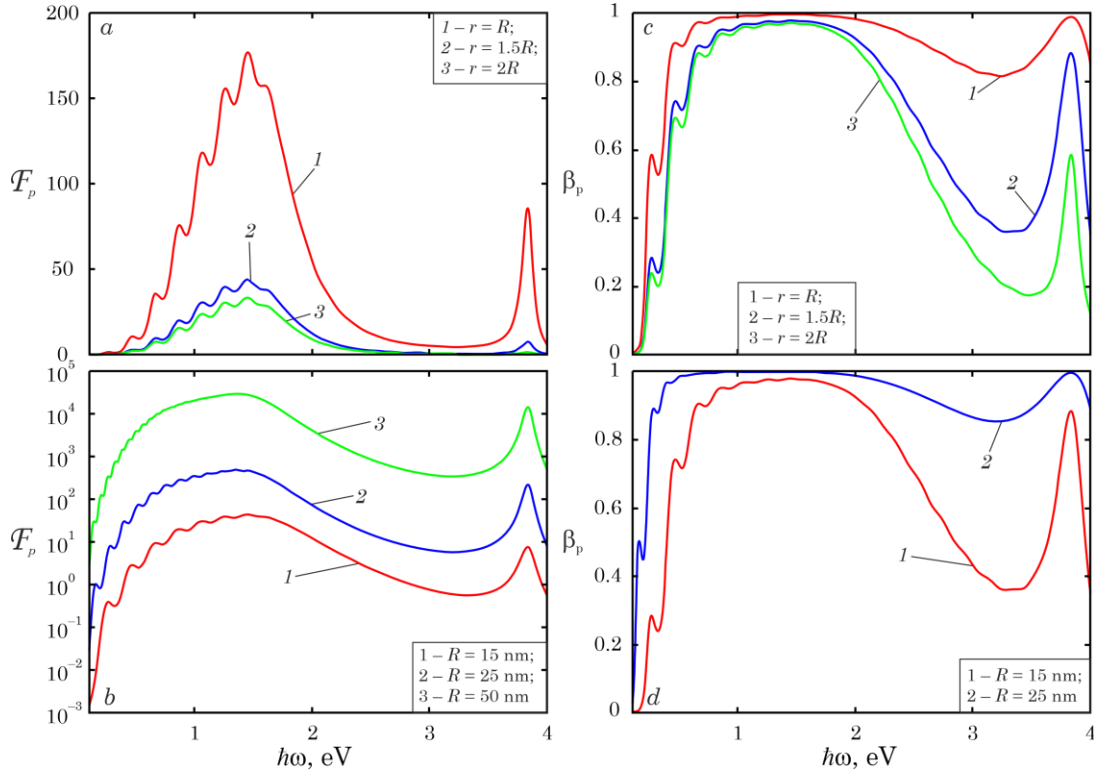
In fig. 2, *a*, the frequency dependences of the Purcell factor are given for different distances of the dipole from the spherical nanoparticle of the radius  $R = 15$  nm at  $\theta = 0^\circ$ . The results of the calculations show a decrease in the Purcell factor with the distance of the dipole molecule from the particle. Two maxima are observed, the presence of which is explained by the following reasons. The first maximum (at a higher frequency) is associated with surface plasmon resonance. The second maximum is due to the resonance of whispering gallery modes of very small mode volume. Small-scale oscillations in the infrared region are a manifestation of kinetic effects.

The magnitude of the Purcell factor under the same other conditions is also significantly affected by the value of the radius of the nanoparticle (Fig. 2, *b*).

In fig. 2, *c*, the frequency dependences of the coupling factor with the nanoparticle-resonator are given for different distances of the dipole from the particle at  $R = 15$  nm,  $\theta = 0^\circ$ . Like the Purcell factor, the coupling factor also increases with decreasing distance from the dipole to the particle. In addition, the closer the dipole is to the particle, the larger the frequency interval in which the coupling factor is near unity. An increase in the radius of nanoparticle also results in an increase in the frequency interval, where  $\beta_p \cong 1$  (Fig. 2, *d*).

We call the dependences of the Purcell factor of the dipole molecule and the coupling factor on the distance  $r$  to the particle, size ones, because this quantity characterizes the size of the molecule-nanoparticle system. The results of the calculations (at  $\omega = \omega_{sp}$ ) of size dependences shown in fig. 3, *a, b* confirm our previous conclusion about the decrease of the Purcell factor and the coupling factor with increasing distance from the dipole to the particle and refine it, demonstrating the monotonicity of these dependencies.

Features of the effect of the nanoparticle material on the frequency and size dependence of the Purcell factor is demonstrated in fig. 4. The shift of maxima of the frequency dependence in Au  $\rightarrow$  Ag  $\rightarrow$  Cu  $\rightarrow$  Pd  $\rightarrow$  Pt series of metals (Fig. 4, *a*)



**Fig. 2** – Frequency dependences of the Purcell factor (*a*, *b*) and the coupling factor with an Ag nanoparticle in air (*c*, *d*) for different values of the distance to the nanoparticle at  $R = 15$  nm (*a*, *c*) and for particles of different radii (*b*, *d*)

is explained by the different values of their optical parameters  $\epsilon^\infty$  and  $\omega_p$ . Curves of size dependence on surface plasmon resonance frequency (Fig. 4, *b*) for nanoparticles of different metals are qualitatively similar. The Purcell factor in all cases gradually decreases with increasing distance between the molecule and the nanoparticle.

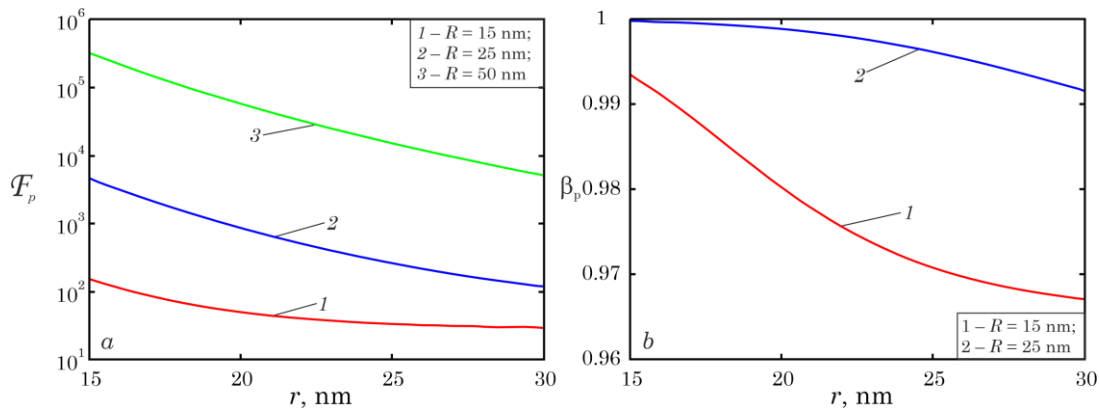
The results of calculations of the frequency and size dependences of the Purcell factor for different environments are shown in fig. 5. Calculations have shown that an increase in dielectric constant in Air  $\rightarrow$  CaF<sub>2</sub>  $\rightarrow$  Teflon  $\rightarrow$  Al<sub>2</sub>O<sub>3</sub>  $\rightarrow$  C<sub>60</sub> series of media results in a decrease in the value of the Purcell factor

while preserving all the previously mentioned features of  $F_p(\hbar\omega)$  and  $F_p(r)$  dependences.

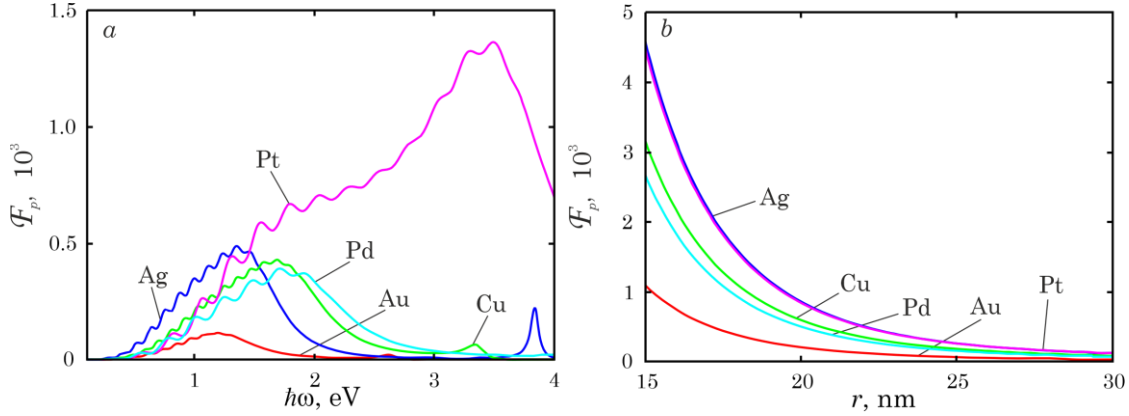
To analyze the shift of the spectral maxima of the Purcell factor, we find them by numerically solving the equation

$$\frac{dF_p}{d\omega} = 0. \tag{19}$$

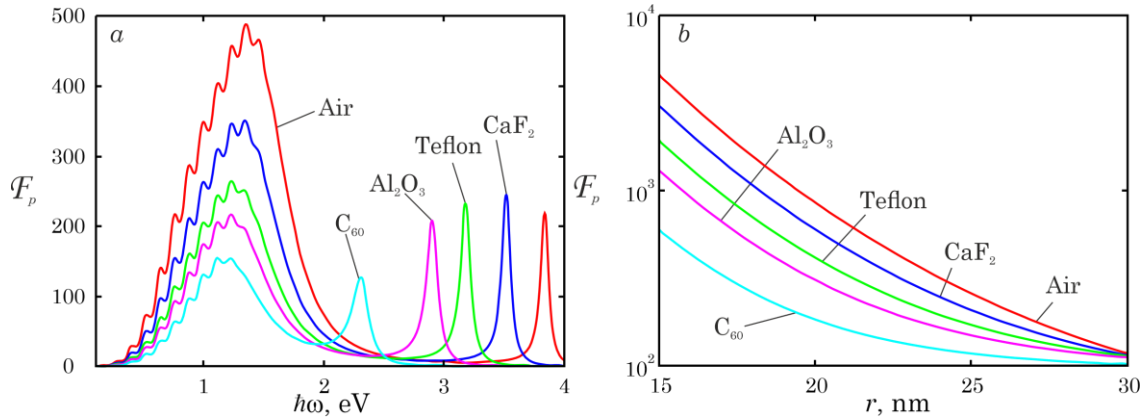
If the dipole molecule and the Ag nanoparticle are in the air ( $\hbar\omega_{sp} = 3.841$  eV) at different distances from each other, we get the results shown in Table 3.



**Fig. 3** – Size dependences of the Purcell factor (*a*) and the coupling factor (*b*) with an Ag nanoparticle in air for different values of the nanoparticle radius



**Fig. 4** – Frequency (a) and size (b) dependences of the Purcell factor of nanoparticles of various metals in air at  $R = 15$  nm



**Fig. 5** – Frequency (a) and size (b) dependences of the Purcell factor of Ag nanoparticles in different dielectric media at  $R = 15$  nm

**Table 3** – Position of the maxima of  $F_p$  at different distances of the dipole molecule from the nanoparticle.

$r$ , nm	15	22.5	30
$\hbar\omega_{\max}$ , eV	1.448	1.446	1.445
	3.836	3.836	3.836

Calculations demonstrate two spectral maxima of the Purcell factor as opposed to one maximum in the frequency dependence of the imaginary part of the polarizability or of absorption cross section of a single spherical Ag nanoparticle. The maximum at a higher frequency actually corresponds to the surface plasmon resonance frequency of an Ag particle in air. Another maximum is observed in the red or infrared region. The shift of both maxima with a change in the distance between the particle and the dipole molecule is practically absent.

A different situation is observed when the radius of the particle increases with the constant distance between the particle and the dipole (Table 4). The maximum at a higher frequency practically does not shift, while the other maximum noticeably shifts towards lower frequencies.

**Table 4** – Position of maxima of  $F_p$  at different nanoparticle radii

$R$ , nm	15	25	50
$\hbar\omega_{\max}$ , eV	1.446	1.348	1.333
	3.836	3.838	3.838

#### 4. CONCLUSIONS

The expressions for the frequency dependences of the Purcell factor and the coupling factor of the molecule-emitter with the nanoparticle-resonator, as well as their size dependencies at the surface plasmon resonance frequency of the nanoparticle, was obtained.

It is shown that the frequency dependences of the Purcell factor and the coupling factor have a maximum at a frequency that practically coincides with the frequency of the surface plasmon resonance, and also another maximum at the resonance frequency of the whispering gallery modes.

As the molecule moves away from the nanoparticle or the radius of the nanoparticle decreases, the values of the Purcell factor and the coupling factor decrease.

The results of calculations for nanoparticles of different metals demonstrate a qualitative similarity of the curves of the frequency and size dependence of the Purcell factor. The quantitative differences are explained by the different value of the plasma frequency and the different fraction of interband transitions in the dielectric function.

It is shown that the surrounding dielectric medium significantly affects the Purcell factor. Thus, for the molecule-Ag nanoparticle system, with an increase in the dielectric constant of the surrounding medium, the Purcell factor decreases along with a decrease in the frequencies at which it reaches maximum values.

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### Фактор Парселла молекули-диполя, розташованої поблизу сферичної металевої наночастинки

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В роботі розглядається задача про вплив сферичної металевої наночастинки на спонтанне випромінювання молекули-диполя, що знаходиться в околі наночастинки. Отримано вирази для частотної і розмірної (на частоті поверхневого плазмонного резонансу) залежностей фактора Парселла, а також фактора зв'язку з резонатором. В якості резонатора виступає наночастинка. Для частотної залежності діелектричної функції вважається придатною модель Друде. Розрахунки проводилися для сферичних наночастинок різного радіусу і з різного металу, розташованих у різних діелектричних середовищах і на різних відстанях до молекули-диполя, з урахуванням об'ємної і поверхневої релаксації та радіаційного загасання.

За результатами розрахунків встановлено, що частотні залежності факторів Парселла та зв'язку з резонатором характеризуються двома максимумами. Один з них, відносно невеликої ширини, спостерігається на частоті, яка практично співпадає з частотою поверхневого плазмонного резонансу поодинокі сферичної металевої наночастинки. Другий, широкий, знаходиться в червоній або інфрачервоній області спектру. Доведено, що причиною виникнення цього другого низькочастотного максимуму є резонанс мод «галереї, що шепоче». За однакової частоти досліджувані фактори мають тим більшу величину, чим більшим є радіус наночастинки і чим меншою є відстань від неї до молекули-диполя. Продемонстровано, що зміна матеріалу наночастинки має наслідком зміну величини фактора Парселла та зсув максимумів в його частотній залежності. Причиною такого ефекту є суттєва відмінність плазмової частоти і частки міжзонних переходів у діелектричній функції для різних металів. Досліджено вплив середовища, в якому знаходиться система срібна наночастинка-диполь, на частотну і розмірну залежність фактора Парселла. Встановлено, що зі збільшенням діелектричної проникності середовища зменшується значення фактора Парселла і максимума його частотної залежності зсуваються в область нижчих частот.

**Ключові слова:** Фактор Парселла, Молекула-диполь, Металева наночастинка, Поверхневий плазмонний резонанс.