Quantum Electronics **35** (10) 891 – 900 (2005)

PHYSICAL BASIS OF QUANTUM ELECTRONICS

PACS numbers: 61.46.+w; 32.50.+d
DOI: 10.1070/QE2005v035n10ABEH013008

Properties of spontaneous radiation of an atom located near a cluster of two spherical nanoparticles

D.V. Guzatov, V.V. Klimov

***Abstract*.** **The analytic solution of the problem of spontaneous decay of an atom near a cluster of two perfectly conducting nanospheres is found. It is shown that spontaneous decay rates can considerably decrease or increase depending on the system geometry. In particular, within the framework of our model, the decay rate of an atom located between closely spaced spheres and having the transition dipole moment directed along the symmetry axis can be arbitrarily high.**

Keywords: *nanoparticles, spontaneous decay rate, nanoclusters.*

1. Introduction

In 1946 Purcell pointed out that the rates of weak radiofrequency atomic transitions can be substantially changed by placing the atom into a resonator [1]. This observation was used in a number of experimental studies of the influence of resonators on the optical properties of atoms [2]. In the last years due to the development of nanotechnologies and methods for manipulating individual atoms and molecules, it was proposed not only to study the optical properties of atoms and molecules located near nanobodies but also to change their properties appropriately for practical purposes. Changes in the properties of emitting atoms located near nanobodies of various shapes and different compositions are used for the development of nanobiosensors [3 – 5], nanolasers [6], artificial fluorophores [7], new efficient light sources [8], efficient and low-cost solar batteries [9], new materials with the negative refractive index [10], microscopes for observation of individual molecules (nanoscopes) [11], instruments for the DNA structure decoding [12], chemical sensors [13 – 15], and many other devices (see analytic review [16]).

From the theoretical point of view, the problem is quite challenging because an atom interacts with strongly inhomogeneous (at the nanometre scale) optical fields. By now the optical properties of atoms near individual nanospheres, nanowires, and nanospheroids have been studied in detail (see reviews [17 – 19]). Very recently the analytic solution of the problem of spontaneous decay rate near a three-axis nanoellipsoid of an arbitrary composition was obtained [7]. However, in all these cases the effect of an individual nanobody on the optical properties of atoms was considered, whereas in practice an atom is located more often near a cluster consisting of a few nanoparticles between which local fields can be considerably stronger, resulting in the enhancement of effects under study [20, 21]. Figure 1 shows clusters consisting of two polystyrene microspheres [22].

D.V. Guzatov, V.V. Klimov P.N. Lebedev Physics Institute, Russian Academy of Sciences, Leninskii prosp. 53, 119991 Moscow, Russia; e-mail: vklim@sci.lebedev.ru

Received 24 June 2005; revision received 3 August 2005
Kvantovaya Elektronika **35** (10) 891 – 900 (2005)
Translated by M.N. Sapozhnikov

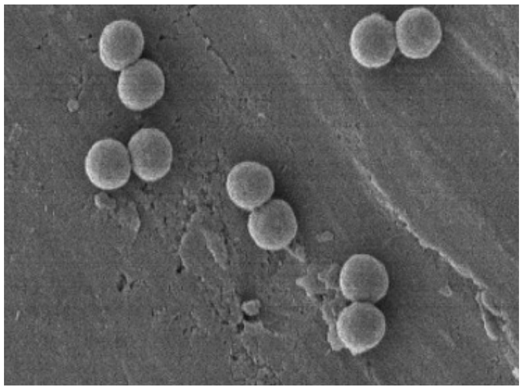

Figure 1. Clusters formed by two spherical nanoparticles [22].

The aim of this paper is to study analytically the optical properties of an atom near a cluster consisting of two perfectly conducting spherical nanoparticles of an arbitrary size and arbitrary mutual disposition (Fig. 2). The optical properties of clusters of two nanoparticles excited by a plane wave have been already studied in papers [23 – 26]. As far as we know, excitation of a two-particle cluster by the dipole radiation of an atom has been studied in the only paper, where a particular case of an atom located exactly midway between nanospheres was considered [27].

2. Elements of the theory of spontaneous emission of an atom near nanobodies

In the case of weak interaction between an atom and a nanobody, i.e., when the atom decays exponentially, the width of the emission line (the decay rate) γ is described by the expression [17, 28, 29]

$$\frac{\gamma}{\gamma_0} = 1 + \frac{3}{2}\,\mathrm{Im}\,\frac{\boldsymbol{d}_0^*\boldsymbol{E}^{\mathrm{r}}(\boldsymbol{r}',\boldsymbol{r}'|\omega_0)}{|d_0|^2 k_0^3}, \tag{1}$$

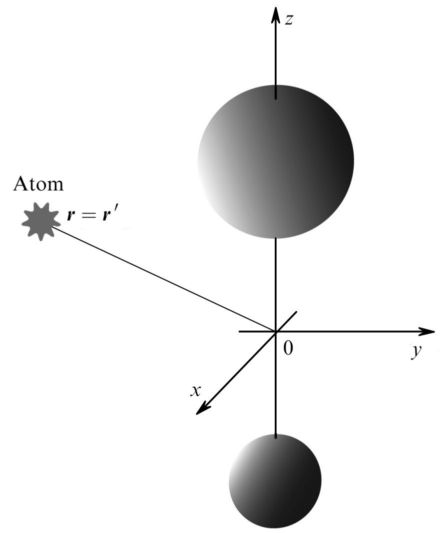

Figure 2. Geometry of the problem.

where $E^{\rm r}_\alpha(\boldsymbol{r}', \boldsymbol{r}'|\omega_0) = G^{\rm r}_{\alpha\beta}(\boldsymbol{r}', \boldsymbol{r}'|\omega_0)d_{0\beta}$ are the components of the reflected field of the dipole $\boldsymbol{d}_0$ near the nanobody at the emission frequency ω_0 of the atom at the atom position $\boldsymbol{r}'$; $G^{\rm r}_{\alpha\beta}$ is the reflected part of the Green function, which can be found from the solution of classical Maxwell's equations with a dipole source; $\alpha, \beta = 1, 2, 3$; γ_0 is the width of the atomic emission line in vacuum; and $k_0 = \omega_0/c$. Note that this expression is valid both for the classical and quantum-mechanical models of an atom. In the quantum-mechanical case, $\boldsymbol{d}_0 = \langle {\rm f}|\hat{\boldsymbol{d}}_0|{\rm i}\rangle$ is the matrix element of the operator of the dipole transition moment of the atom between the initial $|{\rm i}\rangle$ and final $\langle {\rm f}|$ states. When a few decay channels exist, the linewidth is the sum of widths of partial transitions with the corresponding transitions and matrix elements. We will consider below for clearness the case of single-channel decay.

Expression (1) describes the total decay rate of an atom, i.e., both the rate of energy escape to infinity and absorption of energy in a nanobody, and it applicable to bodies made of any materials. Therefore, the study of the spontaneous decay rate of an atom near a nanobody is reduced to the determination of the reflected field and analysis of expression (1). The determination of the reflected field is a challenging problem, which can be solved analytically only in exceptional cases.

For nanobodies, however, the perturbation theory in a small parameter $k_0 b = 2\pi b/\lambda$ can be used, where b is the characteristic size of a nanobody and λ is the radiation wavelength (the Rayleigh theory). In this case, the expression for the reflected field can be expanded as a power series in the wave number k_0:

$$\frac{\boldsymbol{d}_0 \boldsymbol{E}^{\rm r}(\boldsymbol{r}', \boldsymbol{r}'|\omega_0)}{d_0^2} = \tilde{E}_0(\boldsymbol{r}') + \tilde{E}_1(\boldsymbol{r}')k_0 + \tilde{E}_2(\boldsymbol{r}')k_0^2 + {\rm i}\tilde{E}_3(\boldsymbol{r}')k_0^3 + \ldots, \qquad (2)$$

where $\tilde{E}_j(\boldsymbol{r}')$ $(j = 0-3)$ are functions of coordinates, which can be found by solving the corresponding quasi-static problems. The first three terms in the right-hand part of expression (2) describe near-field patterns, whereas the terms beginning from the fourth one describe radiation fields. By substituting expansion (2) into (1), we obtain the total decay rate for an atom near a nanobody

$$\frac{\gamma_{\rm tot}}{\gamma_0} = \frac{3}{2}\,{\rm Im}\left[\frac{\tilde{E}_0(\boldsymbol{r}')}{k_0^3} + \ldots\right] + 1 + \frac{3}{2}\,{\rm Re}\left[\tilde{E}_3(\boldsymbol{r}') + \ldots\right]. \qquad (3)$$

The first term in expression (3) is nonzero only for absorbing media and describes nonradiative losses, while the second and third terms are also nonzero in the absence of absorption and describe radiative losses. Therefore, to determine nonradiative and radiative losses in the first approximation, it is necessary to find $\tilde{E}_0$ and $\tilde{E}_3$, respectively. To find $\tilde{E}_0$, it is sufficient to solve a quasi-static problem with a dipole source. The direct determination of radiative losses described by the terms of third order in k_0 is a difficult problem. However, in the case of an atom located near a nanobody, we are dealing with dipole radiation, and the total dipole moment of the atom + nanobody system can be again found by solving a quasi-static problem in the lowest approximation. Therefore, the radiative linewidth $\gamma_{\rm rad}$ in the case of nanobodies will be described by the expression [30]

$$\frac{\gamma_{\rm rad}}{\gamma_0} = \frac{|\boldsymbol{d}_{\rm tot}|^2}{d_0^2}, \qquad (4)$$

where $\boldsymbol{d}_{\rm tot}$ is the total dipole moment of the atom + nanobody system.

Thus, to find a change in the spontaneous decay rate in the presence of any nanoobject of a size smaller than the radiation wavelength, it is sufficient to solve the quasi-static problem for a dipole near this object.

3. Spontaneous emission of an atom near a cluster consisting of two perfectly conducting spheres

In the case of perfectly conducting spheres, nonradiative losses (absorption) are absent, so that it is sufficient to find only the total dipole moment of the system by solving the quasi-static problem with a dipole source

$$\boldsymbol{E} = -\nabla\psi, \qquad \Delta\psi = 4\pi\exp(-{\rm i}\omega t)(\boldsymbol{d}_0\nabla')\delta^{(3)}(\boldsymbol{r} - \boldsymbol{r}'), \qquad (5)$$

with the corresponding boundary condition on the sphere surface. In the case under study, this condition is the zero tangential components of the field. In expression (5), ∇ and ∇' are the gradients over the coordinates of the observation point $\boldsymbol{r}$ and the atom position $\boldsymbol{r}'$, and ω is the radiation frequency of the atom near the nanobody. To solve Eqn (5), we represent the potential ψ in the form

$$\psi = (\boldsymbol{d}_0\nabla')\tilde{\psi}, \qquad (6)$$

where $\tilde{\psi}$ is the potential of the point unit charge in the presence of two perfectly conducting spheres. It is convenient to solve the Poisson equation

$$\Delta\tilde{\psi} = 4\pi\delta^{(3)}(\boldsymbol{r} - \boldsymbol{r}') \qquad (7)$$

in the bispherical coordinate system (Fig. 3).

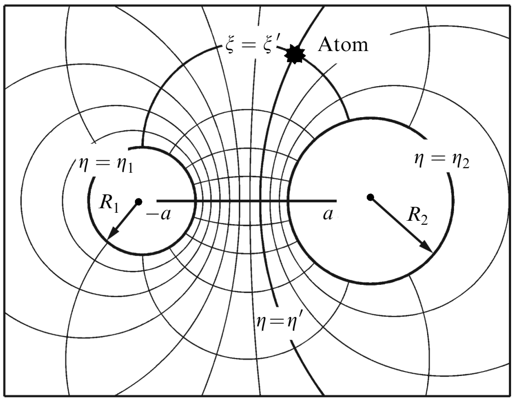

Figure 3. Bispherical coordinate system.

Bispherical coordinates ($-\infty < \eta < \infty$, $0 < \xi \leqslant \pi$, $0 \leqslant \phi < 2\pi$) are related to Cartesian coordinates by the expressions

$$x = a\,\frac{\sin\xi\cos\phi}{\cosh\eta - \cos\xi}, \quad y = a\,\frac{\sin\xi\sin\phi}{\cosh\eta - \cos\xi},$$

$$z = a\,\frac{\sinh\eta}{\cosh\eta - \cos\xi}. \tag{8}$$

The surface $\eta = \eta_1 < 0$ is a sphere of radius $R_1 = a \times |\sinh\eta_1|^{-1}$. Its centre is located at the point $x_1 = y_1 = 0$, $z_1 = a\coth\eta_1$. The second sphere can be specified similarly by the equality $\eta = \eta_2 > 0$. The radius of this sphere is $R_2 = a/\sinh\eta_2$ and the centre is located at the point $x_2 = y_2 = 0$, $z_2 = a\coth\eta_2$. The dimensional constant a is half the distance between the poles of the bispherical coordinate system; it is determined by the positive root of the equation

$$R_{12} = \left(R_1^2 + a^2\right)^{1/2} + \left(R_2^2 + a^2\right)^{1/2},$$

where $R_{12} = z_2 - z_1$ is the distance between the centres of the first and second spheres. Below, the bispherical coordinates of the atom position will be primed.

The solution of Eqn (7) with the specified potentials $\tilde{U}_1$ ($\eta = \eta_1 < 0$) and $\tilde{U}_2$ ($\eta = \eta_2 > 0$) on spheres has the form [31]

$$\tilde{\psi} = \tilde{V} - \sqrt{2}\tilde{U}_1(\cosh\eta - \cos\xi)^{1/2}$$
$$\times\sum_{n=0}^{\infty}\frac{\sinh[(n+1/2)(\eta_2-\eta)]}{\sinh[(n+1/2)(\eta_2-\eta_1)]}\exp\left[\left(n+\frac{1}{2}\right)\eta_1\right]P_n(\cos\xi)$$
$$-\sqrt{2}\tilde{U}_2(\cosh\eta - \cos\xi)^{1/2}\sum_{n=0}^{\infty}\frac{\sinh[(n+1/2)(\eta-\eta_1)]}{\sinh[(n+1/2)(\eta_2-\eta_1)]}$$
$$\times\exp\left[-\left(n+\frac{1}{2}\right)\eta_2\right]P_n(\cos\xi), \tag{9}$$

where

$$\tilde{V} = G_0 + \frac{1}{a}(\cosh\eta - \cos\xi)^{1/2}\sum_{n=0}^{\infty}\sum_{m=0}^{n}P_n^m(\cos\xi)$$
$$\times\left\{A_n\cosh\left[\left(n+\frac{1}{2}\right)\eta\right] + B_n\sinh\left[\left(n+\frac{1}{2}\right)\eta\right]\right\}\times$$
$$\times(c_{mn}\cos m\phi + d_{mn}\sin m\phi); \tag{10}$$

$$A_n = -\frac{\sinh[(n+1/2)\eta_2]\exp[(n+1/2)(\eta_1-\eta')]}{\sinh[(n+1/2)(\eta_2-\eta_1)]}$$
$$-\frac{\sinh[(n+1/2)\eta_1]\exp[-(n+1/2)(\eta_2-\eta')]}{\sinh[(n+1/2)(\eta_2-\eta_1)]};$$
$$B_n = \frac{\cosh[(n+1/2)\eta_2]\exp[(n+1/2)(\eta_1-\eta')]}{\sinh[(n+1/2)(\eta_2-\eta_1)]}$$
$$-\frac{\cosh[(n+1/2)\eta_1]\exp[-(n+1/2)(\eta_2-\eta')]}{\sinh[(n+1/2)(\eta_2-\eta_1)]}; \tag{11}$$

$$G_0(\boldsymbol{r},\boldsymbol{r}') = \frac{1}{|\boldsymbol{r}-\boldsymbol{r}'|} = \frac{1}{a}(\cosh\eta - \cos\xi)^{1/2}$$
$$\times\sum_{n=0}^{\infty}\sum_{m=0}^{n}\exp\left[-\left(n+\frac{1}{2}\right)|\eta-\eta'|\right]P_n^m(\cos\xi)$$
$$\times(c_{mn}\cos m\phi + d_{mn}\sin m\phi) \tag{12}$$

is the Green function of free space in bispherical coordinates [32], in which

$$\left\{\begin{matrix} c_{mn} \\ d_{mn}\end{matrix}\right\} = (2-\delta_{m0})\frac{(n-m)!}{(n+m)!}\left(\cosh\eta' - \cos\xi'\right)^{1/2}$$
$$\times P_n^m\left(\cos\xi'\right)\left\{\begin{matrix}\cos m\phi' \\ \sin m\phi'\end{matrix}\right\}. \tag{13}$$

In our case, the spheres are isolated and their potentials are unknown and are determined by the condition that the total charge Q_i on spheres is zero:

$$Q_i = \frac{a}{4\pi}\int_0^{\pi}\mathrm{d}\xi\,\frac{\sin\xi}{\cosh\eta_i - \cos\xi}\int_0^{2\pi}\mathrm{d}\phi\,\frac{\partial}{\partial\eta}\tilde{\psi}\bigg|_{\eta=\eta_i} = 0 \;\;(i = 1, 2). \tag{14}$$

By calculating integrals (14), we find the potentials of spheres:

$$\tilde{U}_1 = -\frac{(C_{22}+C_{21})\varphi_1 + C_{12}\varphi_2}{C_{12}C_{22} + C_{21}C_{11} + C_{11}C_{22}},$$
$$\tilde{U}_2 = -\frac{C_{21}\varphi_1 + (C_{11}+C_{12})\varphi_2}{C_{12}C_{22} + C_{21}C_{11} + C_{11}C_{22}}, \tag{15}$$

where

$$C_{11} = 2\sum_{n=0}^{\infty}\frac{\sinh[(n+1/2)\eta_2]}{\sinh[(n+1/2)(\eta_2-\eta_1)]}\times$$
$$\times\exp\left[\left(n+\frac{1}{2}\right)\eta_1\right];$$

$$C_{12} = C_{21} = \sum_{n=0}^{\infty}\frac{\exp[-(n+1/2)(\eta_2-\eta_1)]}{\sinh[(n+1/2)(\eta_2-\eta_1)]}; \tag{16}$$

$$C_{22} = -2\sum_{n=0}^{\infty} \frac{\sinh[(n+1/2)\eta_1]}{\sinh[(n+1/2)(\eta_2-\eta_1)]} \times \exp\left[-\left(n+\frac{1}{2}\right)\eta_2\right];$$

$$\varphi_1 = \frac{\sqrt{2}}{a}\sum_{n=0}^{\infty} c_{0n} \frac{\sinh[(n+1/2)(\eta_2-\eta')]}{\sinh[(n+1/2)(\eta_2-\eta_1)]} \times \exp\left[\left(n+\frac{1}{2}\right)\eta_1\right];$$

$$\varphi_2 = \frac{\sqrt{2}}{a}\sum_{n=0}^{\infty} c_{0n} \frac{\sinh[(n+1/2)(\eta'-\eta_1)]}{\sinh[(n+1/2)(\eta_2-\eta_1)]} \times \exp\left[-\left(n+\frac{1}{2}\right)\eta_2\right]; \quad (17)$$

Expressions (10)–(17) determine the solution of problem (7), and the solution of the Poisson equation with a dipole source takes the form (6).

To find spontaneous decay rates (4) from the general solution, it is necessary to determine the dipole moment induced on spheres. This can be done most simply by finding the asymptotics of ψ at large distances. Let R and θ be the coordinates of the spherical coordinate system with the polar z axis. Then, with an accuracy to the second-order-smallness terms ($R \to \infty$), the coordinates of the bispherical system will take the form $\eta \approx 2(a/R)\cos\theta$ and $\xi \approx 2(a/R)\sin\theta$. By substituting these expressions into (9) and expanding the result as a power serried in a small parameter $a/R \ll 1$, we can find the asymptotics of the potential at large distances. By comparing these asymptotics with the known expression for the dipole potential $\boldsymbol{d}_{\text{tot}}\boldsymbol{R}/R^3$, we find expressions for the components of the induced dipole moment vector $\delta\boldsymbol{d}$ of the system under study:

$$\delta d_x = -\sqrt{2}\,a(\boldsymbol{d}_0\nabla')\sum_{n=1}^{\infty} n(n+1)c_{1n}A_n,$$

$$\delta d_y = -\sqrt{2}\,a(\boldsymbol{d}_0\nabla')\sum_{n=1}^{\infty} n(n+1)d_{1n}A_n, \quad (18)$$

$$\delta d_z = 2\sqrt{2}\,a(\boldsymbol{d}_0\nabla')\sum_{n=0}^{\infty}\left(n+\frac{1}{2}\right)c_{0n}B_n + 4a^2U_1\sum_{n=0}^{\infty}\left(n+\frac{1}{2}\right)\frac{\cosh[(n+1/2)\eta_2]}{\sinh[(n+1/2)(\eta_2-\eta_1)]} \times \exp\left[\left(n+\frac{1}{2}\right)\eta_1\right] - 4a^2U_2\sum_{n=0}^{\infty}\left(n+\frac{1}{2}\right) \times \frac{\cosh[(n+1/2)\eta_1]}{\sinh[(n+1/2)(\eta_2-\eta_1)]}\exp\left[-\left(n+\frac{1}{2}\right)\eta_2\right],$$

where $U_{1,2} = (\boldsymbol{d}_0\nabla')\tilde{U}_{1,2}$. The total dipole moment of the system is defined as $\boldsymbol{d}_{\text{tot}} = \boldsymbol{d}_0 + \delta\boldsymbol{d}$.

Thus, expressions (4) and (18) give a completely and explicit solution of the problem of determining the spontaneous decay rate of an excited atom (molecule) located near two perfectly conducting spheres.

4. Asymptotic analysis of important particular cases

The expressions obtained in the previous section are rather cumbersome, and it is difficult to make with their help any certain conclusions about the influence of a cluster on the properties of radiation. In this section, we will use general expressions for a cluster consisting of two identical spheres, which is most important from a practical point of view.

In subsections 4.2, 4.3, and 4.4, the initial and final states of an atom are chosen so that the matrix element of the dipole moment operator is directed along one of the axes of the Cartesian coordinate system. This situation is encountered, for example, if the quantisation axis of the orbital moment is directed along one of the axes, and the initial or final state is the S or P state with the magnetic quantum number $m = 0$.

4.1 General expressions for spontaneous decay rates in the case of identical spheres

In the case of two identical spheres ($-\eta_1 = \eta_2 = \eta_0 > 0$ or $R_1 = R_2 = R_0$), the general expressions for induced dipole moments are somewhat simplified and take the form

$$\delta d_x = 2\sqrt{2}\,a(\boldsymbol{d}_0\nabla')\sum_{n=1}^{\infty} c_{1n}\cosh\left[\left(n+\frac{1}{2}\right)\eta'\right] \times \frac{n(n+1)}{\exp[(2n+1)\eta_0]+1},$$

$$\delta d_y = 2\sqrt{2}\,a(\boldsymbol{d}_0\nabla')\sum_{n=1}^{\infty} d_{1n}\cosh\left[\left(n+\frac{1}{2}\right)\eta'\right] \times \frac{n(n+1)}{\exp[(2n+1)\eta_0]+1}, \quad (19)$$

$$\delta d_z = -4\sqrt{2}\,a(\boldsymbol{d}_0\nabla')\sum_{n=0}^{\infty} c_{0n}\sinh\left[\left(n+\frac{1}{2}\right)\eta'\right] \times \frac{n+1/2}{\exp[(2n+1)\eta_0]-1} + 4a^2(U_1-U_2) \times \sum_{n=0}^{\infty}\frac{n+1/2}{\exp[(2n+1)\eta_0]-1},$$

where

$$U_1 - U_2 = \frac{2\sqrt{2}}{a}\frac{(\boldsymbol{d}_0\nabla')}{2C_{12}+C_{11}} \times \sum_{n=0}^{\infty} c_{0n}\sinh\left[\left(n+\frac{1}{2}\right)\eta'\right]\frac{1}{\exp[(2n+1)\eta_0]-1}; \quad (20)$$

$$C_{11} = \sum_{n=0}^{\infty}\frac{2}{\exp[(2n+1)\eta_0]+1};$$

$$C_{12} = \sum_{n=0}^{\infty}\frac{2}{\exp[(2n+1)\eta_0]-1}. \quad (21)$$

4.2 Spontaneous decay rates of an atom located midway between spheres

Of special interest is a symmetric problem, i.e., the case when an atom is located exactly midway between two identical spheres ($\eta' = 0$ and $\xi' = \pi$). Without loss of generality, we can assume that $\phi' = 0$ ($y' = 0$) and consider two cases of orientation of the dipole moment of the atom.

The dipole moment is oriented along the x axis ($\boldsymbol{d}_0 = d_0\boldsymbol{e}_x$). In this case, it follows from (19)–(21) that the induced dipole moment has the form ($\delta d_y = \delta d_z = 0$)

$$\delta d_x = 8d_0 \sum_{n=1}^{\infty} (-1)^n \frac{n(n+1)}{\exp[(2n+1)\eta_0]+1}. \tag{22}$$

For $\eta_0 \to 0$ (spheres drawing together), this series slowly converges. To sum the series, we transform (22) to the integral (the Watson transformation [33, 34]) over the contour C_1 (Fig. 4)

$$\frac{\delta d_x}{d_0} = -\mathrm{i}4 \oint_{C_1} \frac{\mathrm{d}z}{\sin \pi z} \frac{z(z+1)}{\exp[(2z+1)\eta_0]+1}. \tag{23}$$

Because the integrand in (23) decreases at infinity everywhere except the real axis, we can use the contour $z = \mathrm{i}y$ ($-\infty < y < \infty$) instead of C_1 to calculate sum (22) and the contour C_2 ($z = -1/2 + \mathrm{i}y$) to calculate asymptotics. The contour $z = \mathrm{i}y$ has no singularities, while the contour $z = -1/2 + \mathrm{i}y$ has poles $z_k = -1/2 + \mathrm{i}(\pi/2\eta_0)(2k+1)$ (where $k = 0, \pm 1, \pm 2, \ldots$). As a result, we obtain the expression for the induced dipole moment:

$$\frac{\delta d_x}{d_0} = -1 + \frac{\pi}{\eta_0} \sum_{k=0}^{\infty} \frac{1 + (\pi^2/\eta_0^2)(2k+1)^2}{\cosh\left[(\pi^2/2\eta_0)(2k+1)\right]}. \tag{24}$$

By retaining several first terms in (24), we obtain good asymptotics in the region of small η_0:

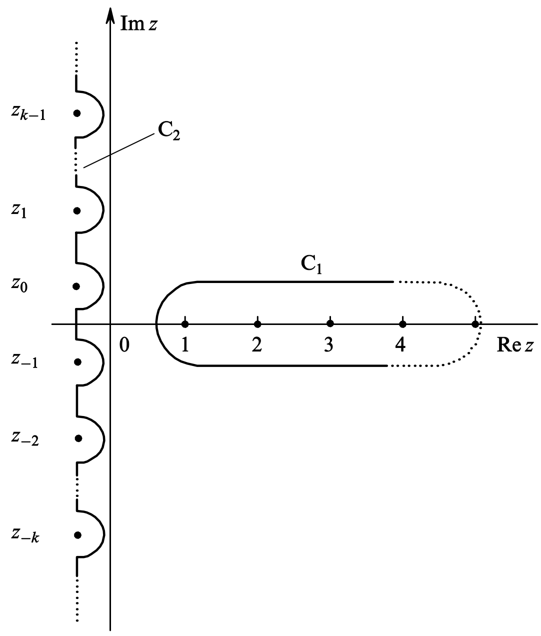

Figure 4. Integration contour in the calculation of asymptotics by the Watson method.

$$\frac{\delta d_x}{d_0} = -1 + 2\frac{\pi^3}{\eta_0^3} \exp\left(-\frac{\pi^2}{2\eta_0}\right) + \ldots. \tag{25}$$

Therefore, the spontaneous emission rate of an atom located exactly midway between spheres and having the dipole moment directed perpendicular to the axis connecting the centres of these spheres proves to be close to zero because the dipole moment induced on spheres completely compensates for the dipole moment of the atom.

The dipole moment is oriented along the z axis ($\boldsymbol{d}_0 = d_0\boldsymbol{e}_z$). In this case, we can obtain from (19)–(21) the expressions ($\delta d_x = \delta d_y = 0$)

$$\delta d_z = -16d_0 \sum_{n=0}^{\infty} (-1)^n \frac{(n+1/2)^2}{\exp[(2n+1)\eta_0]-1} + 4a^2(U_1 - U_2) \sum_{n=0}^{\infty} \frac{n+1/2}{\exp[(2n+1)\eta_0]-1}, \tag{26}$$

where

$$a^2(U_1 - U_2) = 8\frac{d_0}{C_{11}+2C_{12}} \times \sum_{n=0}^{\infty} (-1)^n \frac{n+1/2}{\exp[(2n+1)\eta_0]-1}. \tag{27}$$

In the case of two spheres drawing together, by using the Mellin and Watson transformations (the application of the Mellin transformation to the calculation of asymptotics of series is considered in detail in monograph [31]), we obtain the asymptotics ($\eta_0 \to 0$)

$$\begin{aligned}\frac{\delta d_z}{d_0} \approx {} & -1 + 2\frac{\zeta(2)}{[\gamma_E + \ln(2/\eta_0)]\eta_0^2} + 8\frac{\pi^2}{\eta_0^3} \\ & \times \left[\exp\left(-\frac{\pi^2}{\eta_0}\right) - 4\exp\left(-\frac{2\pi^2}{\eta_0}\right) + \ldots\right] - \frac{1}{6[\gamma_E + \ln(2/\eta_0)]} \\ & \times \left\{1 + \zeta(2) + \frac{\zeta(2)}{6[\gamma_E + \ln(2/\eta_0)]}\right\} + \frac{1}{\gamma_E + \ln(2/\eta_0)} \\ & \times \left\{\frac{1}{72}[\zeta(2)-1] + \frac{1}{\gamma_E + \ln(2/\eta_0)}\left\{\frac{1}{432} + \frac{43}{21600}\zeta(2) \right.\right. \\ & \left.\left. + \frac{\zeta(2)}{2592[\gamma_E + \ln(2/\eta_0)]}\right\}\right\}\eta_0^2 + \ldots,\end{aligned} \tag{28}$$

where ζ is the Riemann zeta function and $\gamma_E \approx 0.577216$ is the Euler constant.

Therefore, the spontaneous emission rate of an atom located exactly midway between spheres and having the dipole moment directed along the straight line connecting the centres of spheres infinitely increases as the nanospheres are drawing together.

4.3 Spontaneous decay rates of an atom located on the surface of one of the spheres

The case of tangential orientation of the dipole moment with respect to the surface is trivial: the boundary conditions are such that the decay rates of an atom become zero. From the physical point of view, this is explained by the fact that the dipole moment induced on spheres is equal to the dipole moment of the atom.

The case of normal orientation of the dipole ($\boldsymbol{d}_0 = d_0\boldsymbol{e}_z$) to the sphere surface is more complicated, and we will consider only the situations when an atom is located at the surface point farthest from the system centre ($\xi' \to 0$) or at the surface point closest to the system centre ($\xi' = \pi$). Without loss of generality, we will assume that the atom is located in the $\phi' = 0$ ($y' = 0$) plane.

For an atom located at the farthest point on the sphere surface ($\eta' = \eta_0$ and $\xi' \to 0$), we can obtain from (19)–(21) the expression for the component of the induced dipole moment:

$$\delta d_z = d_0(\cosh\eta_0 + 2) + 16 d_0 \sinh^3\left(\frac{\eta_0}{2}\right)\sum_{n=0}^{\infty}\left(n+\frac{1}{2}\right)^2 \times \frac{\exp[-(n+1/2)\eta_0]}{\exp[(2n+1)\eta_0]-1} + 4a^2(U_1 - U_2) \times \sum_{n=0}^{\infty}\frac{n+1/2}{\exp[(2n+1)\eta_0]-1}, \quad (29)$$

where

$$a^2(U_1 - U_2) = -\frac{d_0}{C_{11}+2C_{12}}\left\{\sinh\eta_0 + 8\sinh^3\left(\frac{\eta_0}{2}\right) \times \sum_{n=0}^{\infty}\left(n+\frac{1}{2}\right)\frac{\exp[-(n+1/2)\eta_0]}{\exp[(2n+1)\eta_0]-1}\right\}. \quad (30)$$

For an atom located at the surface point closest to the system centre ($\eta' = \eta_0$ and $\xi' = \pi$), we find from (19)–(21) the expression for the component of the induced dipole moment

$$\delta d_z = -d_0(\cosh\eta_0 - 2) - 16 d_0 \cosh^3\left(\frac{\eta_0}{2}\right) \times \sum_{n=0}^{\infty}(-1)^n\left(n+\frac{1}{2}\right)^2\frac{\exp[-(n+1/2)\eta_0]}{\exp[(2n+1)\eta_0]-1} + 4a^2(U_1 - U_2) \times \sum_{n=0}^{\infty}\frac{n+1/2}{\exp[(2n+1)\eta_0]-1}, \quad (31)$$

where

$$a^2(U_1 - U_2) = 8\frac{d_0}{C_{11}+2C_{12}}\cosh^3\left(\frac{\eta_0}{2}\right)\sum_{n=0}^{\infty}(-1)^n \times \left(n+\frac{1}{2}\right)\frac{\exp[(n+1/2)\eta_0]}{\exp[(2n+1)\eta_0]-1}. \quad (32)$$

The rest of the components of the dipole moment are zero in both cases ($\delta d_x = \delta d_y = 0$) due to symmetry considerations.

If spheres are located close to each other ($\eta_0 \to 0$), the series in (29)–(32) converge slowly. In this case, they can be summed by using the Mellin and Watson transformations. As a result, we obtain simple expressions for the induced dipole moment in an important particular case of two closely spaced nanospheres for $\xi' \to 0$ and $\xi' = \pi$, respectively:

$$\frac{\delta d_z}{d_0} \approx -1 + \frac{7}{2}\zeta(3) - \frac{3}{4}\frac{\zeta^2(2)}{\gamma_E + \ln(2/\eta_0)} - 2\frac{\pi^3}{\eta_0^3} \times (8 + 3\eta_0^2 + \ldots)\left[\exp\left(-\frac{\pi^2}{\eta_0}\right) + 4\exp\left(-\frac{2\pi^2}{\eta_0}\right) + \ldots\right] + \frac{1}{16}\left\{7\zeta(3) + \frac{\zeta(2)}{\gamma_E + \ln(2/\eta_0)} \times \left\{1 - \frac{3}{2}\zeta(2) + \frac{\zeta(2)}{6[\gamma_E + \ln(2/\eta_0)]}\right\}\right\}\eta_0^2 + \ldots \quad (33)$$

and

$$\frac{\delta d_z}{d_0} \approx -1 + 2\frac{\zeta(2)}{[\gamma_E + \ln(2/\eta_0)]\eta_0^2} - \frac{1}{6[\gamma_E + \ln(2/\eta_0)]} \times \left\{1 - 5\zeta(2) + \frac{\zeta(2)}{6[\gamma_E + \ln(2/\eta_0)]}\right\} + \frac{1}{\gamma_E + \ln(2/\eta_0)} \times \left\{\frac{1}{72}[11\zeta(2) - 5] + \frac{1}{\gamma_E + \ln(2/\eta_0)} \times \left\{\frac{1}{432} - \frac{257}{21600}\zeta(2) + \frac{\zeta(2)}{2592[\gamma_E + \ln(2/\eta_0)]}\right\}\right\}\eta_0^2 + \ldots. \quad (34)$$

Thus, if an atom has the dipole moment directed along the straight line connecting two perfectly conducting spheres and is itself located on this line (on the surface of one of the spheres), the spontaneous emission rate of the atom can either infinitely increase as the spheres approach each other (if the atom is located on the inner surface of one of the nanospheres) or take finite values (if the atom is located on the external surface of a sphere). In the latter case, the spontaneous decay rate increased by a factor of $[(7/2)\zeta(3)]^2 \approx 17.7$ compared to the decay rate in free space, which is almost twice as large as the increase in the decay rate of an atom located on the surface of an individual sphere, which is 9 times.

In this case, the principal terms in expression (34) for induced dipole moments coincide with the principal terms of the expression for induced dipole moments (28) of an atom located between nanospheres. This coincidence is natural because the positions of the atom considered in the case of an infinitely small distance between nanospheres become indiscernible.

4.4 Widely separated spheres

Of great interest is also the case of spheres separated by distances much greater than the sphere radius. This corresponds to the condition $-\eta_1 = \eta_2 = \eta_0 \to \infty$. By using (19)–(21), we find expressions for induced dipole moments.

The dipole moment is oriented along the x axis ($\boldsymbol{d}_0 = d_0\boldsymbol{e}_x$):

$$\delta d_x \approx 2d_0[2(\cosh\eta' - \cos\xi')]^{1/2}\left[3\sin^2\xi'\cosh\left(\frac{5}{2}\eta'\right) - 2(\cosh\eta' - \cos\xi')\cosh\left(\frac{3}{2}\eta'\right)\right]\exp(-3\eta_0) + \ldots, \quad (35)$$

$$\delta d_z \approx -6d_0\left[2(\cosh\eta' - \cos\xi')\right]^{1/2}\sin\xi'$$

$$\times\left[\sinh\left(\frac{3}{2}\eta'\right) - \cos\xi'\sinh\left(\frac{5}{2}\eta'\right)\right]\exp(-3\eta_0) + \ldots .$$

The dipole moment is oriented along the y axis ($\boldsymbol{d}_0 = d_0\boldsymbol{e}_y$):

$$\delta d_y \approx -2d_0\left[2(\cosh\eta' - \cos\xi')\right]^{3/2} \times\cosh\left(\frac{3}{2}\eta'\right)\exp(-3\eta_0) + \ldots . \quad (36)$$

The dipole moment is oriented along the z axis ($\boldsymbol{d}_0 = d_0\boldsymbol{e}_z$):

$$\delta d_x \approx -6d_0\left[2(\cosh\eta' - \cos\xi')\right]^{1/2}\sin\xi'$$

$$\times\left[\sinh\left(\frac{3}{2}\eta'\right) - \cos\xi'\sinh\left(\frac{5}{2}\eta'\right)\right]\exp(-3\eta_0) + \ldots , \quad (37)$$

$$\delta d_z \approx 2d_0\left[2(\cosh\eta' - \cos\xi')\right]^{1/2}$$

$$\times\left[2\cosh\left(\frac{1}{2}\eta'\right) - 4\cos\xi'\cosh\left(\frac{3}{2}\eta'\right)\right.$$

$$\left.-(1 - 3\cos^2\xi')\cosh\left(\frac{5}{2}\eta'\right)\right]\exp(-3\eta_0) + \ldots .$$

All components of the induced dipole moment that are absent in (35)–(37) are zero.

Expressions (35)–(37) are inadequate if an atom is located on one of the spheres or near it. Consider, for example, an atom located on the sphere surface ($\eta' = \eta_0$). Then, in the case of spheres far removed from each other, the expressions for the nonzero components of the induced dipole moment have the following form:

The dipole moment oriented along the x axis:

$$\frac{\delta d_x}{d_0} \approx 2 - 3\cos^2\xi' + 2\cos\xi'\sin^2\xi'\exp(-\eta_0)$$

$$+ 6(1 + 3\cos 2\xi')\sin^2\xi'\exp(-2\eta_0) + 3(-1 + 4\cos\xi'$$

$$+ 8\cos 3\xi')\sin^2\xi'\exp(-3\eta_0) + \ldots , \quad (38)$$

$$\frac{\delta d_z}{d_0} \approx \frac{3}{2}\sin 2\xi' - 3(\sin\xi' - \sin 3\xi')\exp(-\eta_0)$$

$$-3\left(2\sin 2\xi' - \frac{3}{2}\sin 4\xi'\right)\exp(-2\eta_0) + 3(\sin\xi' + \sin 2\xi'$$

$$- 3\sin 3\xi' + 2\sin 5\xi')\exp(-3\eta_0) + \ldots .$$

The dipole moment oriented along the y axis:

$$\delta d_y = -d_0. \quad (39)$$

The dipole moment is oriented along the z axis:

$$\frac{\delta d_x}{d_0} \approx \frac{3}{2}\sin 2\xi' + 6\cos 2\xi'\sin\xi'\exp(-\eta_0) -$$

$$-3\left(2\sin 2\xi' - \frac{3}{2}\sin 4\xi'\right)\exp(-2\eta_0) + 3\left(\sin\xi' - \frac{1}{2}\sin 2\xi'\right.$$

$$\left.- 3\sin 3\xi' + 2\sin 5\xi'\right)\exp(-3\eta_0) + \ldots , \quad (40)$$

$$\frac{\delta d_z}{d_0} \approx -1 + 3\cos^2\xi' - 12\cos 2\xi'\sin^2\xi'\exp(-\eta_0)$$

$$- 6(1 + 3\cos 2\xi')\sin^2\xi'\exp(-2\eta_0) + 3(1 + \cos\xi' + \cos 2\xi'$$

$$- 3\cos 3\xi' + 2\cos 5\xi')\exp(-3\eta_0) + \ldots .$$

By retaining the principal terms in these expressions, we see that, depending on the position of the atom, we can obtain different spontaneous decay rates for different orientations of the atomic dipole moment. Thus, assuming that $\xi' = 0$ ($\xi' = \pi$) or $\xi' = \pi/2$, we obtain the relative spontaneous decay rate equal to 9 (if the dipole moment of the atom is oriented along the normal to the sphere surface) or zero (if the dipole moment is oriented perpendicular to the normal to the surface). This completely agrees with the solution of the problem on the spontaneous emission of an atom located near a single sphere [17, 18].

Expressions for the induced dipole moment of two widely separated identical spheres can be also obtained without using bispherical coordinates by replacing spheres by point dipoles with polarisability equal to the polarisability of a sphere in a homogeneous field. This replacement is correct because in this case the field in the vicinity of spheres is almost homogeneous. We assume that an atom is located at a point with the coordinate $\boldsymbol{r}_0$ and has the dipole moment $\boldsymbol{d}_0$. Let us denote the dipole moments of nanospheres and radius vectors of their centres as $\boldsymbol{d}_1$, $\boldsymbol{d}_2$ and $\boldsymbol{r}_1$, $\boldsymbol{r}_2$, respectively, and assume that the radii of nanospheres are $R_1 = R_2 = R_0$. Then, the self-consistent system of equations for dipole moments has the form

$$\boldsymbol{d}_1 = \varkappa[\boldsymbol{E}_2(\boldsymbol{r}_1) + \boldsymbol{E}_0(\boldsymbol{r}_1)],$$
$$\boldsymbol{d}_2 = \varkappa[\boldsymbol{E}_1(\boldsymbol{r}_2) + \boldsymbol{E}_0(\boldsymbol{r}_2)], \quad (41)$$

where

$$\boldsymbol{E}_i(\boldsymbol{r}) = -\frac{\boldsymbol{d}_i}{|\boldsymbol{r} - \boldsymbol{r}_i|^3} + 3\frac{(\boldsymbol{d}_i(\boldsymbol{r} - \boldsymbol{r}_i))(\boldsymbol{r} - \boldsymbol{r}_i)}{|\boldsymbol{r} - \boldsymbol{r}_i|^5} \quad (i = 0, 1, 2). \quad (42)$$

The polarisability of a perfectly conducting sphere is $\varkappa = R_0^3$. The total dipole moment of this system is $\boldsymbol{d}_{\rm tot} = \boldsymbol{d}_0 + \boldsymbol{d}_1 + \boldsymbol{d}_2$, and the spontaneous emission rate is determined by expression (4). The solution of system (41) can be found quite easily if the z axis is taken as the rotation symmetry axis and it is assumed that $-z_1 = z_2 = R_{12}/2$. Then, the nonzero components of the induced dipole moment for an atom located in the $y' = 0$ plane can be written in the following form:

The dipole moment is oriented along the x axis:

$$\delta d_x = d_0\left(\frac{1}{R_0^3} + \frac{1}{R_{12}^3}\right)^{-1}\left(-\frac{1}{R_-^3} + 3\frac{x'^2}{R_-^5} - \frac{1}{R_+^3} + 3\frac{x'^2}{R_+^5}\right),$$
$$\delta d_z = 3d_0\left(\frac{1}{R_0^3} - \frac{2}{R_{12}^3}\right)^{-1}\left(\frac{z' - R_{12}/2}{R_-^5} + \frac{z' + R_{12}/2}{R_+^5}\right)x'. \quad (43)$$

The dipole moment is oriented along the y axis:

$$\delta d_y = -d_0\left(\frac{1}{R_0^3}+\frac{1}{R_{12}^3}\right)^{-1}\left(\frac{1}{R_-^3}+\frac{1}{R_+^3}\right). \quad (44)$$

The dipole moment is oriented along the z axis:

$$\delta d_x = 3d_0\left(\frac{1}{R_0^3}+\frac{1}{R_{12}^3}\right)^{-1}\left(\frac{z'-R_{12}/2}{R_-^5}+\frac{z'+R_{12}/2}{R_+^5}\right)x',$$

$$\delta d_z = d_0\left(\frac{1}{R_0^3}-\frac{2}{R_{12}^3}\right)^{-1} \quad (45)$$

$$\times\left[-\frac{1}{R_-^3}+3\,\frac{(z'-R_{12}/2)^2}{R_-^5}-\frac{1}{R_+^3}+3\,\frac{(z'+R_{12}/2)^2}{R_+^5}\right],$$

where

$$R_\pm = \left[x'^2+\left(z'\pm\frac{1}{2}R_{12}\right)^2\right]^{1/2}.$$

By expanding expressions (43)–(45) in a series for $R_0 \to 0$, we can obtain the same expressions as (35)–(40). A distinct feature of expressions (43)–(45) is that they are valid for any position of the atom with respect to spheres, including on their surface. This is explained by the fact that the dipole moment of a nanospheres in the quasi-stationary regime is always equal to the product of polarisability on the dipole-moment field of the atom at the sphere centre [7].

5. Graphic illustrations and discussion of results

In this section, we will consider as an example a cluster consisting of two identical nanospheres of radius 50 nm. Figure 5 demonstrates the spontaneous decay rate of an atom located in the $y' = 0$ plane. It is assumed that the atom can be located at any point of this plane, and the value of spontaneous decay rate at this point is shown by different shades of grey. The direction of the dipole moment of the atom is shown by a standard symbol: the arrow or circle.

One can easily see that the decay rate takes the maximum value on the surface of one of the spheres at the point where the direction of the dipole moment coincides with that of the normal to the surface sphere (Fig. 5a). In this case, the spontaneous decay rate is maximal if the atom is located on the straight line connecting the centres of nanospheres, on the inner side of the sphere. The spontaneous decay rate takes the minimum value (zero) when the dipole is located on the sphere surface and the dipole moment is directed perpendicular to the normal to the sphere surface (Fig. 5b). As nanospheres approach each other, the spontaneous decay rate increases infinitely if the atom is located on the rotation axis between spheres and has the dipole moment oriented along this axis (Fig. 5c) [see expressions (34) and (28)] or tends to zero if the dipole moment is directed perpendicular to the rotation axis and is located on it in the region between spheres (Figs 5a, b).

Figure 6 shows the change in the spontaneous decay rate when the atom moves away from the system of nanospheres. As expected, the relative spontaneous decay rate at large distances from the cluster tends to unity, i.e., to the value corresponding to the spontaneous decay rate of an atom (molecule) in vacuum in the absence of a nanoobject. Note

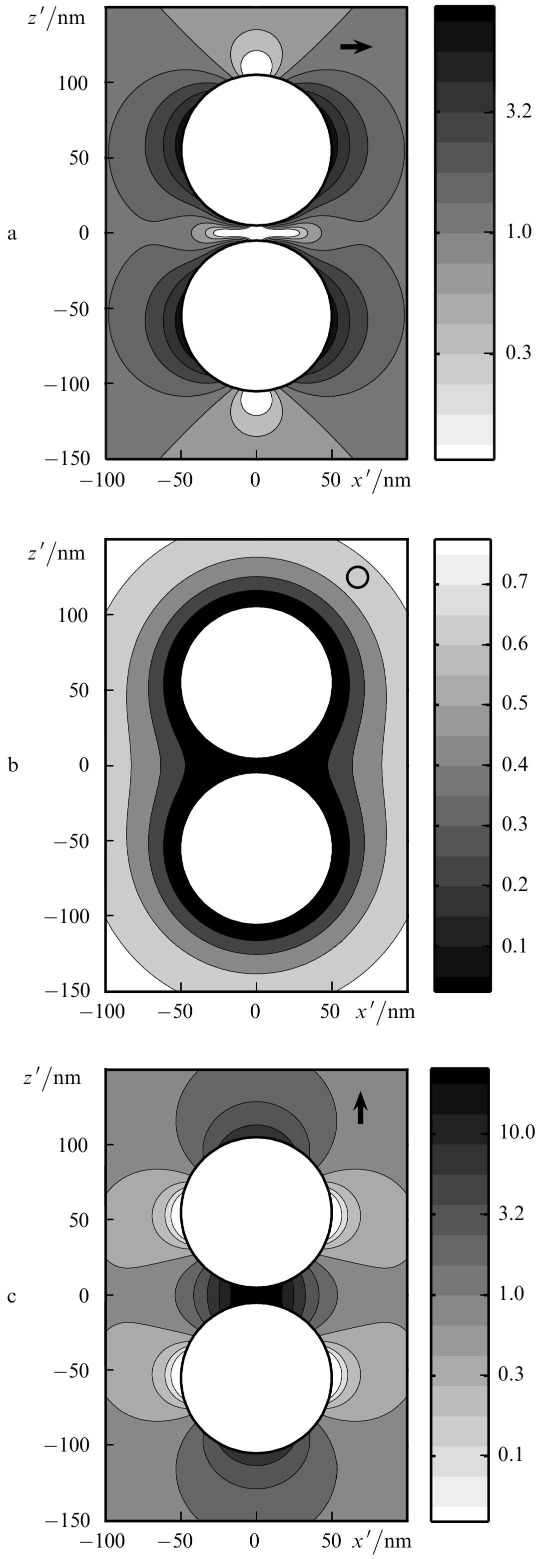

Figure 5. Spontaneous decay rates of an atom (molecule) in the $y' = 0$ plane for the dipole moment oriented along axes x (a), y (b), and z (c). The radii of spheres are 50 nm and the distance between their centres is 110 nm.

that the atom with the z-orientation of the dipole moment near the cluster has a high spontaneous decay rate [curve (*3*) in Fig. 6a].

Figure 7 shows the decay rate of the atom located at the characteristic points of the system as a function of the

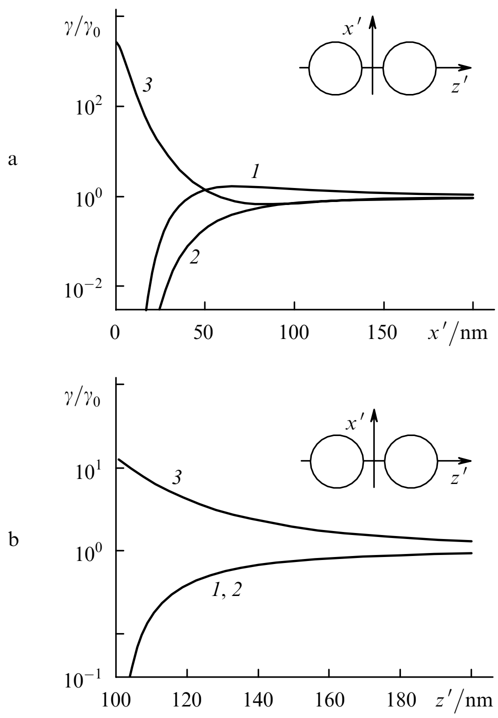

Figure 6. Spontaneous decay rate of an atom (molecule) as a function of the distance between the atom and the system of nanospheres. The spheres have radii of 50 nm and the distance between their centers is 101 nm. The atom moves away from the exact center between the spheres along the x axis (a) and from the external surface of one of the spheres along the z axis (b). The dipole moment is oriented along x (*1*), y (*2*) and z (*3*) axes.

distance between spheres and the corresponding asymptotics. One can see again that the decay rate of the atom with the dipole moment oriented along the z axis and located inside the cluster (Figs 7a, b) noticeably increases. It also follows from Fig. 7 that asymptotic expressions (25), (28), and (33), (34) and also (35)–(40) [or (43)–(45)] describe the entire range of variation of the spontaneous decay rate of an atom near two spheres.

6. Conclusions

We have studied the spontaneous decay of an atom near a cluster consisting of two perfectly conducting nanospheres. Analytic expressions have been found for the decay rates in the form of series for an arbitrary geometry of the system. Simpler expressions were obtained for a cluster consisting of two identical spheres. It is shown that in the case of closely spaced spheres, the spontaneous decay rates can infinitely increase for an atom located between the spheres and having the dipole moment directed along the symmetry axis.

The analytic equations obtained in the paper can be used not only for prompt estimates of the effect of clusters on fluorescence of atoms and molecules but also for precision estimates of numerical calculations in more complicated cases.

The calculation method developed in this paper can be generalised to a cluster consisting of two spheres made of arbitrary materials. In this case, of course, the values of decay rates change substantially compared to the case of ideally conducting spheres considered here. The study of the

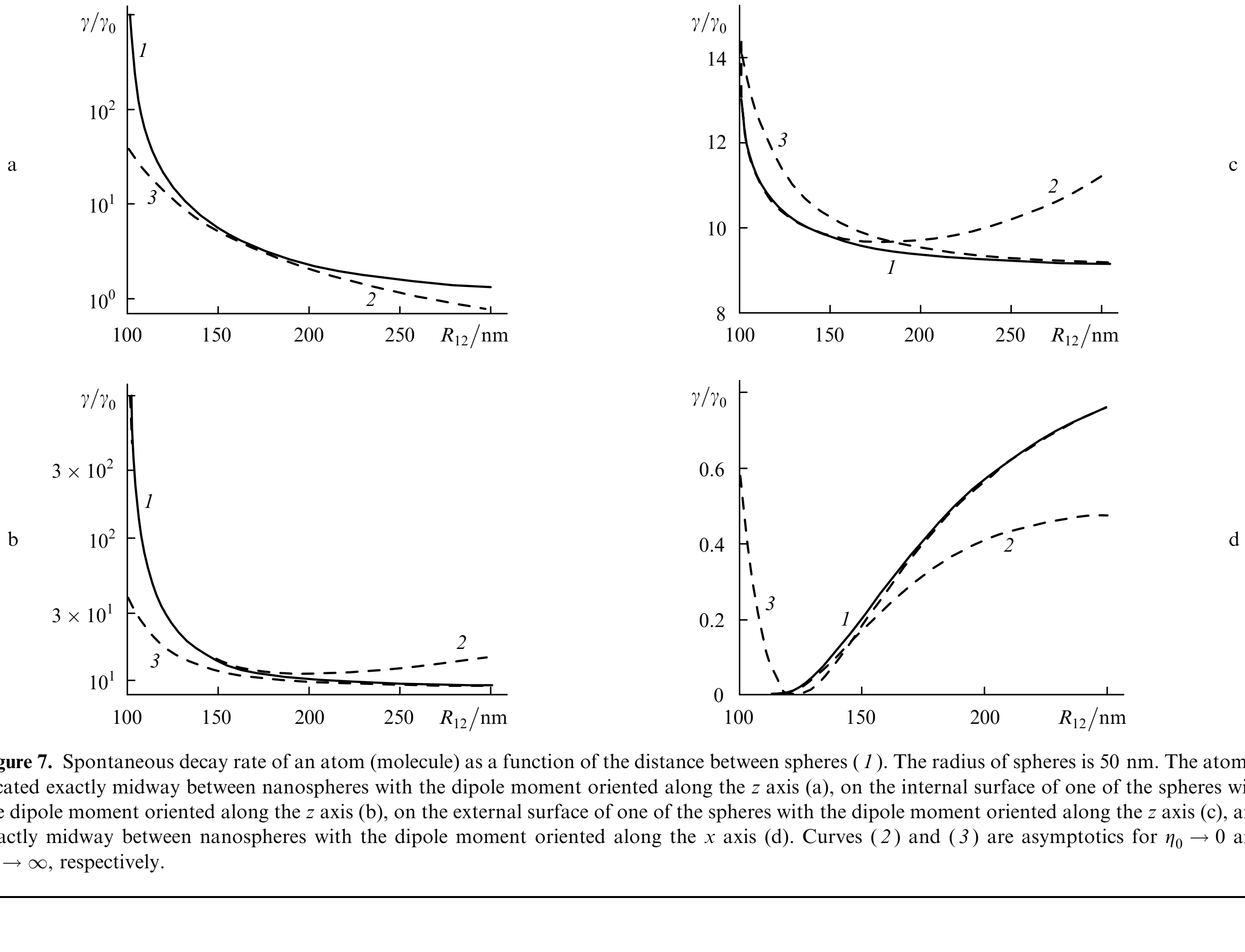

Figure 7. Spontaneous decay rate of an atom (molecule) as a function of the distance between spheres (*1*). The radius of spheres is 50 nm. The atom is located exactly midway between nanospheres with the dipole moment oriented along the z axis (a), on the internal surface of one of the spheres with the dipole moment oriented along the z axis (b), on the external surface of one of the spheres with the dipole moment oriented along the z axis (c), and exactly midway between nanospheres with the dipole moment oriented along the x axis (d). Curves (*2*) and (*3*) are asymptotics for $\eta_0 \to 0$ and $\eta_0 \to \infty$, respectively.

spontaneous emission of an atom located near more realistic metal and dielectric clusters will be presented elsewhere.

Acknowledgements. This work was partially supported by the Russian Foundation for Basic Research (Grant No. 04-02-16211) and the Program of the Presidium of RAS 'Effect of Atomic Crystal and Electronic Structure on the Properties of Condensed Media'.

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