Quadrupole radiation of an atom in the vicinity of a dielectric microsphere

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The process of quadrupole radiation of an atom in the vicinity of a dielectric sphere is considered within the framework of both quantum-mechanical and classical approaches. It is shown that spontaneous transition probabilities can be calculated correctly within the framework of the classical approach. The quadrupole transition probability is shown to be capable of increasing by several orders of magnitude in the neighborhood of the microsphere, to become comparable with the intensity of dipole transitions, the frequency shifts calculated in the classical approximation being much greater than in the case of dipole transitions. [S1050-2947(96)06410-4]

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I. INTRODUCTION

It is well known that the rates of quadrupole transitions in the optical region are lower by a factor of $(a_0/\lambda)^2 \propto 10^{-6}-10^{-8}$ (where a_0 is the Bohr radius and λ is the radiation wavelength) than those of their dipole counterparts and that the dipole transition probability is strongly influenced by the presence of macroscopic bodies near the radiating atom (see, for example, [1,2]. The question arises in this connection: How do material bodies affect quadrupole or multipole transitions? The effect of a dielectric sphere on the linewidth of a quadrupole transition was considered in [3]. This paper, a sequel to [3], presents the results of investigations into this question, which show that, given appropriate conditions, the probability of quadrupole or other multipole transitions can rise substantially, and that atomic transition frequencies suffer material shifts in the neighborhood of a surface.

Consider the amplitude of the decay of an excited atomic state to an unexcited (metastable) state, accompanied by the emission of a photon. In that case, the transition matrix element has the form

$$V_{fi} \propto \int \psi_{\mathrm{out}}^*(\mathbf{r}) \boldsymbol{\nabla} \psi_{\mathrm{in}}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r},\omega) d^3\mathbf{r}, \tag{1.1}$$

where $\mathbf{A}$ is the wave function of the photon emitted, with allowance made for the presence of material bodies.

As in the case of free space, the wave functions of an atom vary faster than the wave function of the photon, and this allows one to expand the wave function of the photon into a series in powers of coordinates in the vicinity of the atom.

Where dipole radiation is forbidden, the first term in this series goes to zero, and the value of the matrix element is governed by that of the gradient of the photon wave function in the neighborhood of the atom,

$$V_{fi} \propto \frac{\partial}{\partial r_{0,j}} \mathbf{A}_i(\mathbf{r}_0,\omega) \int \psi_{\mathrm{out}}^*(\mathbf{r}) \frac{\partial}{\partial r_i} \psi_{\mathrm{in}}(\mathbf{r}) r_j d^3\mathbf{r}. \tag{1.2}$$

A principal difference between the case in hand and that of free space is that the scale of the gradient of the photon wave function depends, generally speaking, not only on the radiation wavelength but also on the characteristic size of the problem. What is more, in the case of an atom located close to a material body with a small radius of curvature, a, the wave-function gradient is determined mainly by the surface curvature of the body and not by the radiation wavelength in free space. As a result, the quadrupole radiation probability increases $(\lambda/a)^2$ times as compared with that in the case of free space. An even greater increase in probability should be expected for multipole transitions of higher order. Note the fact that where the characteristic geometrical size of the problem is close to the size of the atomic orbit, the radiation intensity may approach the intensity of dipole transitions. Specifically, for Rydberg and closely similar atoms, the orbit size may be as great as 10^{-5} cm, and so one can create appropriate geometrical conditions for the observation of the enhancement of quadrupole transitions.

The plan for the rest of the paper is as follows. In Sec. II, we find, within the framework of the lower-order perturbation theory approximation (as to charge), quantum-mechanical expressions for the probability of spontaneous quadrupole decay of an atom located next to a dielectric sphere. In Sec. III, in the classical nonresonance approximation we treat the radiation of a multipole in the vicinity of a dielectric sphere, and find expressions for the radiation linewidth and frequency shift. In conclusion, we compare between results obtained within the frameworks of the classical and quantum-mechanical approaches and, examine these results.

II. LINEWIDTH OF A QUADRUPOLE TRANSITION (QUANTUM-MECHANICAL APPROACH)

For the sake of definiteness consider an electric quadrupole transition of an atom located close to a dielectric sphere

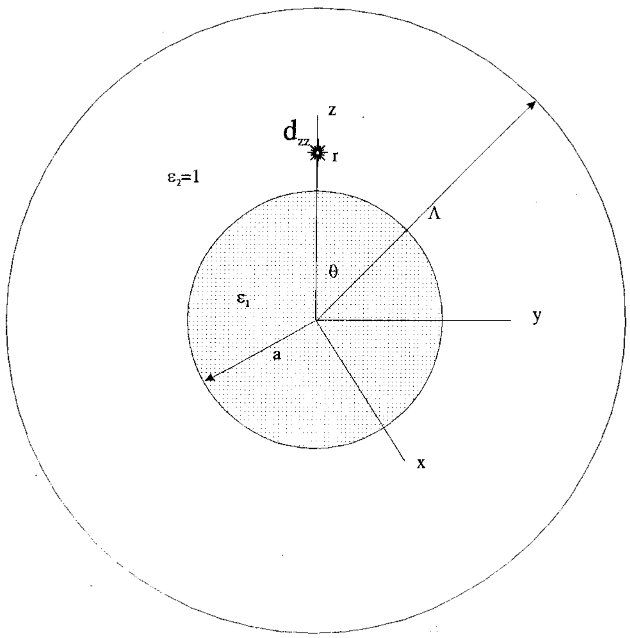


FIG. 1. Geometry of the quantum-mechanical problem.

whose radius is small compared to the radiation wavelength (see Fig. 1 for the geometry of the problem). Other transitions and problem geometries will be analyzed in a separate publication.

A. Quantization of electromagnetic field in the presence of a dielectric microsphere

The quantization procedure is generally known quite well, but each particular case requires a special approach. In our case, an ideally conducting sphere of a great but finite radius $\Lambda\to\propto$ (see Fig. 1) may be treated as the quantization volume. The expansion of the electromagnetic field and its vector potential into a series in the complete set of eigenfunctions of the classical problem (see, for example, [4]) may be represented in the form

$$\mathbf{E}=\sum_s \frac{a_s-a_s^\dagger}{i\sqrt{2}}\,\mathbf{e}(s,\mathbf{r});\mathbf{B}=\sum_s \frac{a_s+a_s^\dagger}{\sqrt{2}}\,\mathbf{b}(s,\mathbf{r}),$$

$$\mathbf{A}=-\frac{c}{\omega_s}\sum_s \frac{a_s+a_s^\dagger}{\sqrt{2}}\,\mathbf{e}(s,\mathbf{r}), \tag{2.1}$$

$$\boldsymbol{\nabla}\times\mathbf{e}(s,\mathbf{r})=-\frac{\omega_s}{c}\,\mathbf{b}(s,\mathbf{r}).$$

Here $\mathbf{a}_s$ and $\mathbf{a}_s^\dagger$ are the coefficients of photon annihilation and creation in the appropriate modes with the frequencies ω_s.

In the case of an electric quadrupole transition, both TM and TE waves can be excited, depending on the orientation of the atom, only the TM waves being excited in the case of radial arrangement of the atomic quadrupole momentum. The expressions for the electric field $\mathbf{e}(s,\mathbf{r})$ of these modes can easily be obtained in terms of spherical harmonics (Y) and spherical Bessel functions (j) and Hankel functions (h) [5]. For the fields outside the sphere that are of interest to us, the following expressions hold:

$$\mathbf{e}_{\mathrm{TE}}(n,m,\nu)=[\alpha_{\mathrm{TE},n}^{(1)}h_n^{(1)}(k_2r)+\alpha_{\mathrm{TE},n}^{(2)}h_n^{(2)}(k_2r)]\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi), \tag{2.2}$$

$$\mathbf{e}_{\mathrm{TM}}(n,m,\nu)=-\frac{1}{k_2}\,\boldsymbol{\nabla}\times\{[\alpha_{\mathrm{TM},n}^{(1)}h_n^{(1)}(k_2r)+\alpha_{\mathrm{TM},n}^{(2)}h_n^{(2)}\times(k_2r)]\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi)\}.$$

Here $\hat{\mathbf{L}}=-i(\mathbf{r}\times\boldsymbol{\nabla})$ is the angular momentum operator, n is the orbital quantum number, m is the azimuthal quantum number, ν is the radial quantum number, and $k_{1,2}=\sqrt{\varepsilon_{1,2}}k=\sqrt{\varepsilon_{1,2}}(\omega_s/c)$ are the wave vectors within and without the sphere, respectively.

The coefficients α are found as usual such that the tangential field components at the sphere boundary, are continuous and the wave functions are normalized within the sphere of radius Λ per photon in a mode:

$$\frac{\alpha_{\mathrm{TE}}^{(1)}}{\alpha_{\mathrm{TE}}^{(2)}}=1-2p_n\,,\quad \frac{\alpha_{\mathrm{TM}}^{(1)}}{\alpha_{\mathrm{TM}}^{(2)}}=1-2q_n\,,$$

$$q_n=\frac{\left[\varepsilon\,\frac{d}{dz_2}\,[z_2j_n(z_2)]j_n(z_1)-\frac{d}{dz_1}\,[z_1j_n(z_1)]j_n(z_2)\right]}{\left[\varepsilon\,\frac{d}{dz_2}\,[z_2h_n^{(1)}(z_2)]j_n(z_1)-\frac{d}{dz_1}\,[z_1j_n(z_1)]h_n^{(1)}(z_2)\right]}, \tag{2.3}$$

$$p_n=\frac{\left[\frac{d}{dz_2}\,[z_2j_n(z_2)]j_n(z_1)-\frac{d}{dz_1}\,[z_1j_n(z_1)]j_n(z_2)\right]}{\left[\frac{d}{dz_2}\,[z_2h_n^{(1)}(z_2)]j_n(z_1)-\frac{d}{dz_1}\,[z_1j_n(z_1)]h_n^{(1)}(z_2)\right]},$$

$$|\alpha_{\mathrm{TE},n}^{(1)}|^2=|\alpha_{\mathrm{TE},n}^{(2)}|^2=|\alpha_{\mathrm{TM},n}^{(1)}|^2=|\alpha_{\mathrm{TM},n}^{(2)}|^2=\frac{2\pi\hbar c}{\Lambda}\,\frac{k^3}{n(n+1)}.$$

In expression (2.3) and elsewhere, $z_{1,2}=k_{1,2}a$.

To find the total probability, one must also know the density of final states. The requirement that the tangential electric-field components of the TM mode should vanish on the inside surface of the normalization sphere gives rise to the following transcendental equation:

$$\left.\frac{d}{dr}\,(rZ)\right|_{r=\Lambda}=0,$$
$$Z=\left[\alpha_{\mathrm{TM},n}^{(1)}h_n^{(1)}\left(\frac{\omega_s}{c}\,r\right)+\alpha_{\mathrm{TM},n}^{(2)}h_n^{(2)}\left(\frac{\omega_s}{c}\,r\right)\right], \tag{2.4}$$

whose asymptotic solutions are

$$\omega_s=\left(\nu+\frac{n+1}{2}\right)\frac{\pi c}{\Lambda}+\cdots\,. \tag{2.5}$$

Hence it follows that the density of final states will be described by the simple expression

$$\rho_{\mathrm{TM}}(\omega)=\frac{\Lambda}{\pi\hbar c}. \tag{2.6}$$

Analyzed in a similar way are those TE photons for which the density of final states is expressed in exactly the same way:

$$\rho_{\mathrm{TE}}(\omega)=\frac{\Lambda}{\pi\hbar c}. \tag{2.7}$$

B. Spontaneous quadrupole transition linewidth

According to the golden rule (see, for example, [6]), the probability and linewidth of a transition are given by

$$\gamma=\frac{2\pi}{\hbar}\,|V_{fi}|^2\rho(\omega), \tag{2.8}$$

so that the problem is reduced to the finding of the transition matrix element.

Within the framework of the low-order perturbation theory as to charge, the interaction energy matrix element has the form

$$\langle \mathrm{out}|V_{\mathrm{int}}|\mathrm{in}\rangle=\left\langle \mathrm{out}\left|\,\frac{e}{c}\int d^3r\hat{\mathbf{j}}\cdot\hat{\mathbf{A}}\right|\mathrm{in}\right\rangle$$
$$=-\frac{i\hbar e}{\sqrt{2m\omega_s}}\int d^3r\ \psi^*_{\mathrm{out}}\nabla\psi_{\mathrm{in}}\cdot\mathbf{e}(s,\mathbf{r}), \tag{2.9}$$

where $\mathbf{e}(s,\mathbf{r})$ is the electric field of the appropriate mode. In our case, the dipole transition is forbidden (or is of no interest to us), so that the quadrupole transition amplitude has the form

$$V_{fi}=-\frac{i\hbar e}{\sqrt{2m\omega_s}}\frac{\partial}{\partial r_{0,j}}A_i(\mathbf{r}_0,\omega)\int \psi^*_{\mathrm{out}}(\mathbf{r})\frac{\partial}{\partial r_i}\psi_{\mathrm{in}}(\mathbf{r})r_j d^3\mathbf{r}, \tag{2.10}$$

where $\mathbf{r}_0$ is the atomic position vector.

Of course, to use the perturbation theory as to charge, one should consider the fact that in the region near to resonance, the matrix element may become large enough, the smallness of the charge notwithstanding. It was demonstrated in [7] that for microspheres of a not very great size in comparison with the wavelength, the first-order perturbation theory approximation for dipole transitions remains applicable even in the vicinity of resonance. The quadrupole interaction being weaker than its dipole counterpart, the first-order perturbation theory approximation also proves to be correct for quadrupole transitions in the case of small spheres ($ka\sim1$). This question will be considered in more detail in an individual publication.

To find the gradient of the wave functions in (2.10) in the neighborhood of the emitting atom, it is convenient to use a spherical coordinate system and make the covariant differentiation of the vectors [8] in this system. As a result, the expression for the transition matrix element assumes the form

$$V_{fi}=\frac{Q_{ij}D_{ij}}{6\sqrt{2}}. \tag{2.11}$$

Here $D_{ij}=e\langle(3x_ix_j-\delta_{ij})\rangle_{fi}$ is the transition quadrupole moment, and $Q_{ij}=\nabla_i e^j$ is the covariant derivative of the electric field of the appropriate mode [8]:

$$\nabla_i e^j=\frac{1}{H_j}\frac{\partial e^j}{\partial x^i}-\frac{e^i}{H_j^2}\frac{\partial H^i}{\partial x^i}+\delta_i^j\sum_{k=1}^{3}\frac{e^k}{H_k}\frac{\partial\ln H^j}{\partial x^k}, \tag{2.12}$$

where $H_i=(1,r,r\sin\theta)_i$ are the Lame coefficients of the spherical coordinate system and use is made on the right-hand side of the physical (not covariant) coordinates of the electric-field vectors.

In deriving expression (2.11), use has been made of the following relation for the matrix elements:

$$\left\langle f\left|x_i\frac{\partial}{\partial x_j}\right|i\right\rangle=\frac{m\omega_{fi}}{2\hbar}\langle f|x_ix_j|i\rangle, \tag{2.13}$$

which holds true for quadrupole transitions involving a change in the principal or the orbital quantum number, as well as the condition for the field to be solenoidal, $Q_{ij}=\mathrm{div}\,\mathbf{e}(s,\mathbf{r})=0$. Here $\omega_{fi}=\omega_s$ is the transition frequency.

Expression (2.11) is valid for any quadrupole orientation. Let us now consider some particular cases of quadrupole orientation. In the case of radial orientation of the principal dipole axis, i.e., where the quadrupole momentum tensor (in the spherical coordinate system) has the form

$$D_{ij}=\begin{bmatrix} D_{rr} & 0 & 0\\ 0 & -\frac{1}{2}D_{rr} & 0\\ 0 & 0 & -\frac{1}{2}D_{rr}\end{bmatrix}_{ij}, \tag{2.14}$$

the expression for the matrix element assumes the form

$$V_{fi}=\frac{Q_{rr}D_{rr}}{4\sqrt{2}}, \tag{2.15}$$

where the radial component (r) of the covariant derivative has the simple form

$$Q_{rr}=\frac{\partial e_r}{\partial r}, \tag{2.16}$$

and no summation is extended over the terms with repeated indices. Note that in the case of radial quadrupole orientation, there takes place no emission of TE photons, and so one should use e_r as only the second expression in (2.2).

Substituting (2.15), (2.16), and (2.6) into (2.8), we obtain the final expression for the total quadrupole transition probability for a radially oriented quadrupole in the vicinity of a dielectric sphere [3]:

$$\gamma^Q_{\mathrm{TM}}=\frac{D_{rr}^2k^5}{8\hbar}\sum_{n=1}^{\infty}n(n+1)(2n+1)\times\left|\frac{d}{dz}\left(\frac{j_n(z)-q_nh_n^{(1)}(z)}{z}\right)\right|^2, \tag{2.17}$$

where $z=ka$.

Relating this expression to the spontaneous decay rate in a vacuum (see, for example, [9]),

$$\gamma_{\rm vac}^{Q}=\frac{D_{zz}^2 k^5}{60\hbar}, \tag{2.18}$$

we obtain the final expression for the relative spontaneous quadrupole transition rate for a radially oriented quadrupole in the neighborhood of a dielectric sphere:

$$\frac{\gamma_{\rm rad}^{Q}}{\gamma_{\rm vac}^{Q}}=\frac{15}{2}\sum_{n=1}^{\infty} n(n+1)(2n+1)\left|\frac{d}{dz}\left(\frac{j_n(z)-q_n h_n^{(1)}(z)}{z}\right)\right|^2, \tag{2.19}$$

In the case of tangential orientation of the principal dipole axis, i.e., where the quadrupole moment tensor (in the spherical coordinate system) has the form

$$D_{ij}=\begin{bmatrix} -\frac{1}{2}D_{\vartheta\vartheta} & 0 & 0 \\ 0 & D_{\vartheta\vartheta} & 0 \\ 0 & 0 & -\frac{1}{2}D_{\vartheta\vartheta}\end{bmatrix}_{ij}, \tag{2.20}$$

the expression for the matrix element assumes the form

$$V_{fi}=\frac{Q_{\theta\theta}D_{\theta\theta}}{4\sqrt{2}}, \tag{2.21}$$

where the tangential component (θ) of the covariant derivative has the form

$$Q_{\theta\theta}=\frac{1}{r}\frac{\partial e_{\vartheta}}{\partial\vartheta}+\frac{e_r}{r}, \tag{2.22}$$

and no summation is extended over the terms with repeated indices. Note that in the case of tangential orientation of the principal dipole axis, there occurs emission of both TM and TE photons, and therefore one should take as e_r and e_θ both the first and the second expression in (2.2).

Substituting (2.21), (2.22), and (2.6) into (2.8), we obtain the final expression for the total quadrupole transition probability for a tangentially oriented quadrupole in the vicinity of a dielectric sphere:

$$\begin{aligned}\gamma_{\rm tan}^{Q}=&\frac{D_{\theta\theta}^2 k^5}{32\hbar}\sum_{n=1}^{\infty} n(n+1)(2n+1)\left|\frac{d}{dz}\left(\frac{j_n(z)-q_n h_n^{(1)}(z)}{z}\right)\right|^2\\ &+\frac{D_{\theta\theta}^2 k^5}{64\hbar}\sum_{n=1}^{\infty} n(n-1)(n+2)\\ &\times\left|\frac{1}{z^2}\frac{d}{dz}\{z[j_n(z)-q_n h_n^{(1)}(z)]\}\right|^2\\ &+\frac{D_{\theta\theta}^2 k^5}{64\hbar}\sum_{n=1}^{\infty} n(n-1)(n+2)\left|\frac{1}{z}[j_n(z)-p_n h_n^{(1)}(z)]\right|^2\end{aligned} \tag{2.23}$$

where $z=ka$.

Relating this expression to the spontaneous decay rate in a vacuum (17), we obtain the final relation for the relative spontaneous quadrupole transition rate for a tangentially oriented dipole in the presence of a dielectric microsphere:

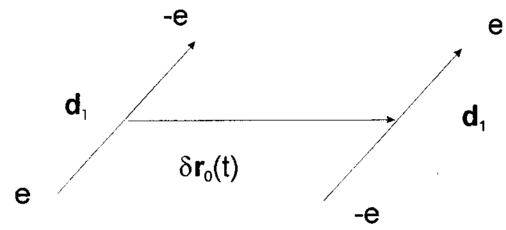


FIG. 2. Geometry of the classical problem.

$$\begin{aligned}\frac{\gamma_{\rm tan}^{Q}}{\gamma_{\rm vac}^{Q}}=&\frac{15}{8}\sum_{n=1}^{\infty} n(n+1)(2n+1)\left|\frac{d}{dz}\left(\frac{j_n(z)-q_n h_n^{(1)}(z)}{z}\right)\right|^2\\ &+\frac{15}{16}\sum_{n=1}^{\infty}(2n+1)(n-1)(n+2)\\ &\times\left|\frac{1}{z^2}\frac{d}{dz}\{z[j_n(z)-q_n h_n^{(1)}(z)]\}\right|^2\\ &+\frac{15}{16}\sum_{n=1}^{\infty}(2n+1)(n-1)(n+2)\\ &\times\left|\frac{1}{z}[j_n(z)-p_n h_n^{(1)}(z)]\right|^2.\end{aligned} \tag{2.24}$$

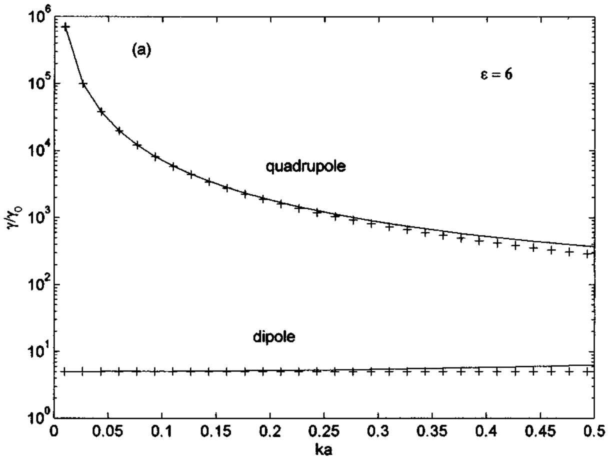
A quadrupole with its principal axis oriented along the φ axis is analyzed in a similar way. As one would expect, the result is an expression identical to (2.23), which is one more argument in support of the correctness of our calculations.

Thus, in the present section, we have found, within the framework of the first-order quantum-electrodynamic perturbation theory, the relative changes that the linewidth of a spontaneous atomic quadrupole radiation suffers in the vicinity of a dielectric sphere, no matter what the orientation of the transition quadrupole moment.

III. LINEWIDTH AND FREQUENCY SHIFT OF A QUADRUPOLE TRANSITION (CLASSICAL APPROACH)

It is a well-established fact that in the case of dipole transitions there exists a simple correspondence between the transition linewidths calculated within the framework of the classical and the quantum-mechanical approaches. That is, the changes occurring in the classical and quantum-mechanical linewidths in the presence of a material body, taken in relation to their counterparts in free space, are described by identical expressions. The natural question arises: Does a similar correspondence exist for quadrupole transitions, too?

To clear up this question, let us consider a system of two dipoles of opposite orientations with the moments ed_1 and $-ed_1$, one of which is stationary and the other, displaced for a distance of $\delta\mathbf{r}(t)$, which oscillates about the first, the system being placed close to a sphere of radius a with a dielectric constant of ε in an infinite medium with a dielectric constant of $\varepsilon=1$. The geometry of the classical problem is shown in Fig. 2. The equation of motion of the movable



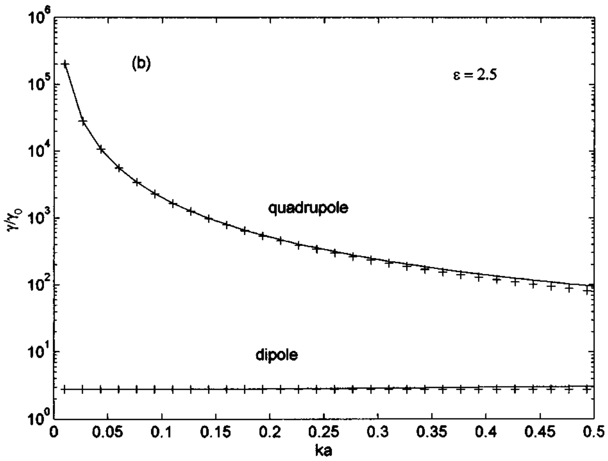


FIG. 3. Relative linewidth for quadrupole and dipole transitions in atoms located in close proximity to the surface of a dielectric microsphere in the case of radial orientation as a function of the microsphere radius ka: (a) $\varepsilon=6$; (b) $\varepsilon=2.5$. The crosses indicate asymptotic relations (4.7) and (4.9).

dipole portion of the quadrupole in the case of weak radiation reaction has the form

$$m\,\delta\ddot{\mathbf{r}}+m\,\gamma_0\,\delta\dot{\mathbf{r}}+m\,\omega_0^2\,\delta\mathbf{r}=\mathbf{0}, \tag{3.1}$$

$$\gamma_0=\frac{2e^2d_1^2}{15c}\,\frac{k^4}{m}=\frac{cD_0^2\bar{\bar{K}}^6}{60E_0}. \tag{3.2}$$

Here γ_0 is the total quadrupole transition width in a vacuum, $D_0=-4e\,\delta r_0 d_1$ is the maximum quadrupole moment of the system, and $E_0=m\omega^2\,\delta r_0^2/2$ is the total initial oscillation energy of the quadrupole.

An oscillating quadrupole located (at r') near to a dielectric sphere is acted upon additionally (compared to the case of free space) by the reflected field $\mathbf{E}^{(1)}(\mathbf{r}')$, so that the equation of motion assumes the form

$$m\,\delta\ddot{\mathbf{r}}_0+m\,\gamma_0\,\delta\dot{\mathbf{r}}_0+m\,\omega_0^2\,\delta\mathbf{r}_0=-e(\mathbf{d}_1\cdot\boldsymbol{\nabla})\mathbf{E}^{(1)}(\mathbf{r}'), \tag{3.3}$$

where $\delta\mathbf{r}_0$ is the oscillation amplitude of the dipole. Projecting this equation onto the oscillation direction, we obtain

$$m\ddot{D}+m\,\gamma_0\dot{D}+m\,\omega_0^2D=4e^2d_1(\mathbf{d}_1\cdot\boldsymbol{\nabla})\,\frac{\delta\mathbf{r}\cdot\mathbf{E}^{(1)}(\mathbf{r}')}{|\delta\mathbf{r}|}, \tag{3.4}$$

where D is the quadrupole moment in the oscillation direction, which is assumed later in the text to be coincident with the principal axis of the quadrupole.

By solving in accordance with the perturbation theory the dispersion equation following from (3.3), one can easily find the formulas for both the linewidth variation,

$$\gamma=\gamma_0-\frac{e}{\omega_0 m\,\delta r_0^2}\,\mathrm{Im}(\mathbf{d}_1\cdot\boldsymbol{\nabla})(\delta\mathbf{r}_0\cdot\mathbf{E}^{(1)}) \tag{3.5}$$

and the frequency shift

$$\Delta\omega=\frac{e}{2\,\omega_0 m\,\delta r_0^2}\,\mathrm{Re}(\mathbf{d}_1\cdot\boldsymbol{\nabla})(\delta\mathbf{r}_0\cdot\mathbf{E}^{(1)}). \tag{3.6}$$

In the case of resonance interaction between the quadrupole and the dielectric sphere, use of the perturbation theory is restricted, as in the quantum-mechanical case, by the requirement that the linewidth should be much smaller than the resonance width of the dielectric sphere. Where the sphere is small, this condition is satisfied for characteristic quadrupole transitions.

Using Eq. (3.1), one can write the following expressions for the relative quantities:

$$\frac{\gamma}{\gamma_0}=1-\frac{15}{2e\,\delta r_0^2d_1^2k^5}\,\mathrm{Im}(\mathbf{d}_1\cdot\boldsymbol{\nabla})(\delta\mathbf{r}_0\cdot\mathbf{E}) \tag{3.7}$$

and

$$\frac{\Delta\omega}{\gamma_0}=\frac{15}{4e\,\delta r_0^2d_1^2k^5}\,\mathrm{Re}(\mathbf{d}_1\cdot\boldsymbol{\nabla})(\delta\mathbf{r}_0\cdot\mathbf{E}). \tag{3.8}$$

Thus, to obtain concrete results, one should calculate $(\mathbf{d}_1\cdot\boldsymbol{\nabla})(\delta\mathbf{r}_0\cdot\mathbf{E})$ at the location of the oscillating dipole.

To find the variation of the quadrupole radiation parameters at any point in space, it is necessary to solve the Maxwell equations for the system of charges under analysis. The charge density of the stationary dipole may be defined by the expression

$$\rho_1=-e(\mathbf{d}_1\cdot\boldsymbol{\nabla})\,\delta(\mathbf{r}-\mathbf{r}'), \tag{3.9}$$

and that of the oscillating dipole by the expression

$$\rho_2=e(\mathbf{d}_1\cdot\boldsymbol{\nabla})\,\delta[\mathbf{r}-\mathbf{r}'-\delta\mathbf{r}(t)]. \tag{3.10}$$

Accordingly, the total charge and current densities may be defined by the expressions

$$\rho_{\mathrm{tot}}=-e(\mathbf{d}_1\cdot\boldsymbol{\nabla})(\delta\mathbf{r}\cdot\boldsymbol{\nabla})\,\delta(\mathbf{r}-\mathbf{r}') \tag{3.11}$$

and

$$\mathbf{j}=e\mathbf{v}(t)(\mathbf{d}_1\cdot\boldsymbol{\nabla})\,\delta(\mathbf{r}-\mathbf{r}'),\quad \mathbf{v}(t)=\delta\dot{\mathbf{r}}(t). \tag{3.12}$$

To solve the Maxwell equations involving the sources defined by expressions (3.10) and (3.11), let us use the method proposed in [11]. To this end, we expand the field of the

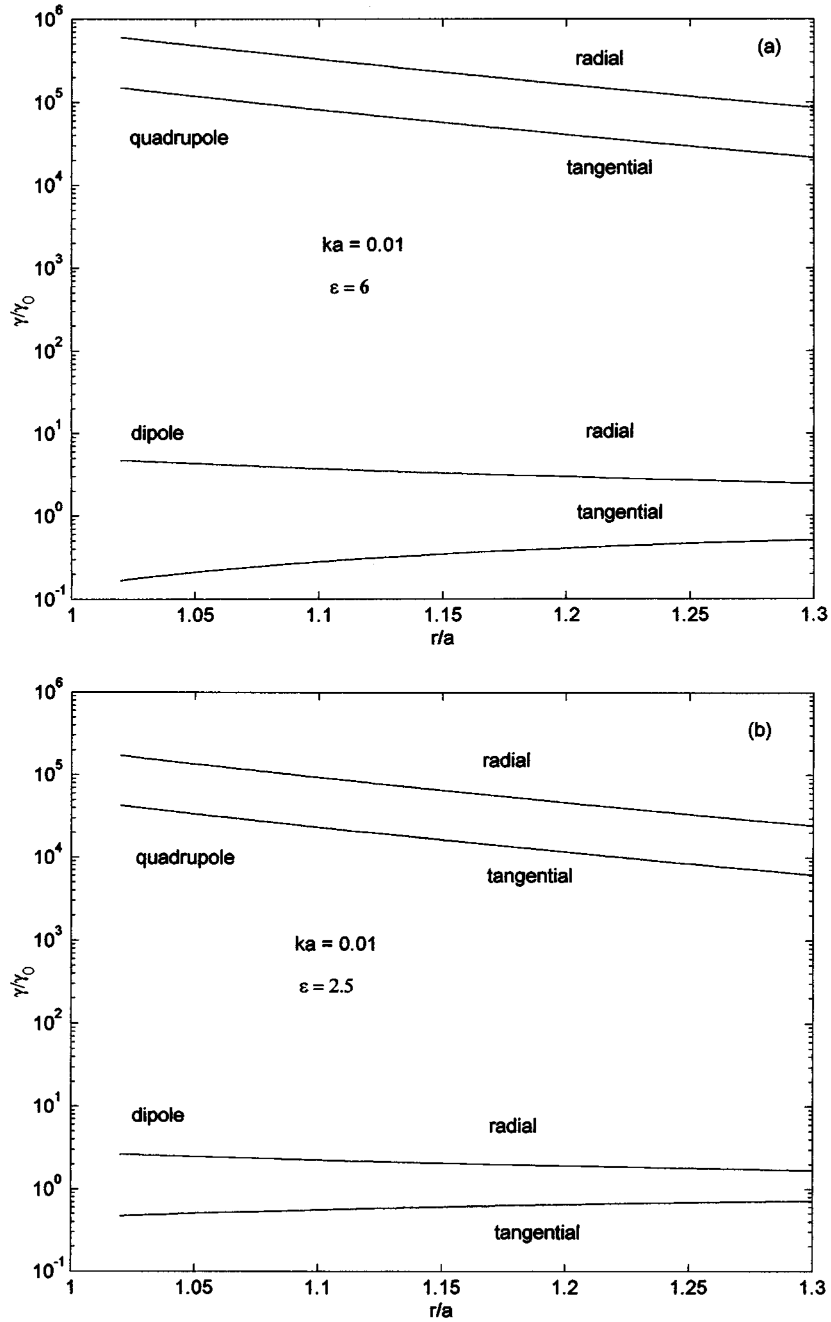


FIG. 4. Linewidth of quadrupole and dipole transitions for various orientations and various refractive indices of the microsphere as a function of the distance r/a to the surface of the microsphere [$ka=0.01$, $\varepsilon=6$ (diamond), $\varepsilon=2.5$ (glass)].

oscillating quadrupole in terms of spherical harmonics, and then find by the ordinary procedure the reflected field [or, to be more exact, the quantity $(\mathbf{d}_1\cdot\boldsymbol{\nabla})(\delta\mathbf{r}_0\cdot\mathbf{E})$] of interest to us.

If current distributions (3.10) and (3.11) are placed in an infinite dielectric sphere with a dielectric constant of $\varepsilon_2=1$, the expression for the field may be written in the form

$$\mathbf{E}^{\mathrm{quadr}}=k^2\boldsymbol{\Pi}^{(1)}+\boldsymbol{\nabla}(\boldsymbol{\nabla}\cdot\boldsymbol{\Pi}^{(1)}), \tag{3.13}$$

$$\mathbf{B}^{\mathrm{quadr}}=-ik\boldsymbol{\nabla}\times\boldsymbol{\Pi}^{(1)}, \tag{3.14}$$

where the Hertzian electric vector of the quadrupole is defined by the expression

$$\boldsymbol{\Pi}^{(1)}=e\,\delta\mathbf{r}(\mathbf{d}_1\cdot\boldsymbol{\nabla})\,\frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}. \tag{3.15}$$

To find the expansion of the quadrupole field in terms of spherical harmonics, let us first consider the radial magnetic-field component. It is not very difficult to demonstrate that there holds true the following important identity:

$$\begin{aligned}(\mathbf{r}\mathbf{B}^{\mathrm{quadr}})&=-ike[\mathbf{r}\cdot(\boldsymbol{\nabla}\times\delta\mathbf{r}_0)](\mathbf{d}_1\cdot\boldsymbol{\nabla})\,\frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}\\&=-ike(\mathbf{d}_1\cdot\boldsymbol{\nabla}')[\delta\mathbf{r}_0\cdot(\boldsymbol{\nabla}'\times\mathbf{r}')]\,\frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}.\end{aligned} \tag{3.16}$$

Thereafter, by using the standard expansion [4]

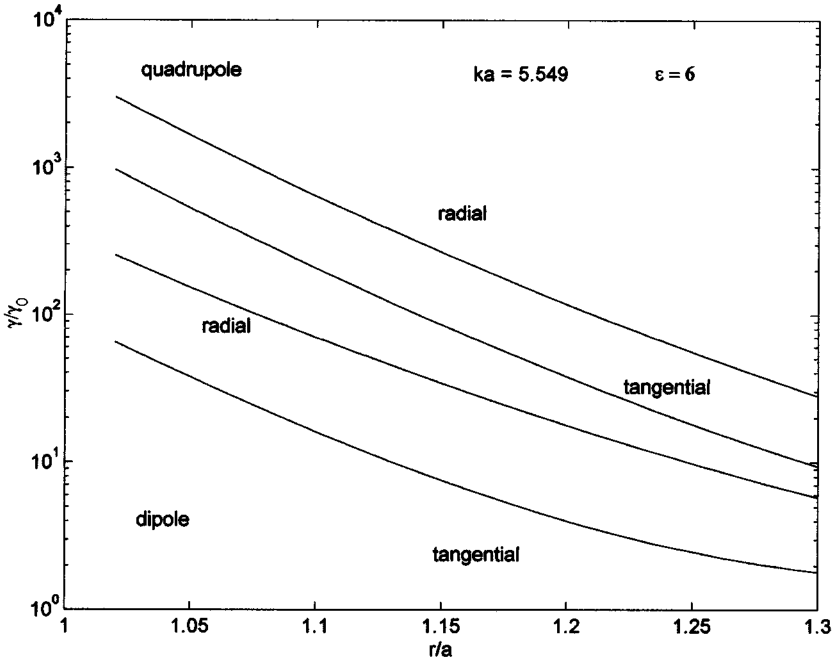


FIG. 5. Linewidth of quadrupole and dipole transitions for various orientations in the case of resonance interaction with a dielectric sphere of a sufficiently large radius ($ka = 5.5491$) compared to the radiation wavelength as a function of the distance r/a to the surface of the microsphere.

$$\frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|} = 4\pi ki \sum_{n=0}^{\infty} \sum_{m=-n}^{n} j_n(kr) h_n^{(1)}(kr') \times Y_{nm}^*(\vartheta',\varphi') Y_{nm}(\vartheta,\varphi), \tag{3.17}$$

which is valid for $r < r'$, the expression for the radial magnetic-field component is reduced to the necessary form of expansion in terms of scalar spherical harmonics:

$$(\mathbf{r}\cdot\mathbf{B}^{\mathrm{quadr}}) = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} a_{nm} j_n(kr) Y_{nm}(\vartheta,\varphi), \tag{3.18}$$

where

$$a_{nm} = ie4\pi k^2 (\mathbf{d}_1\cdot\nabla')(\delta\mathbf{r}_0\cdot\hat{\mathbf{L}}') h_n^{(1)}(kr') Y_{nm}^*(\vartheta',\varphi') \tag{3.19}$$

and $k_{1,2} = k_0\sqrt{\varepsilon_{1,2}}$ are the wave vectors in the regions 1 and 2, respectively. Use is also made here of the ordinary expressions for the spherical harmonics and spherical Bessel functions [4].

Now multiplying the Maxwell equation

$$\nabla\times\mathbf{E} = ik\mathbf{B} \tag{3.20}$$

by $\mathbf{r}$, we obtain the following equation that can help find the TE component of the dipole field:

$$(\hat{\mathbf{L}}\cdot\mathbf{E}_{\mathrm{TE}}^{\mathrm{quadr}}) = k(\mathbf{B}^{\mathrm{quadr}}\cdot\mathbf{r}). \tag{3.21}$$

Solving this equation, we find the TE component of the electric field ($r < r'$) to be

$$\mathbf{E}_{\mathrm{TE}}^{\mathrm{quadr}} = k \sum_{n=1}^{\infty} \sum_{m=-n}^{n} \frac{a_{nm}}{n(n+1)} j_n(kr) \hat{\mathbf{L}} Y_{nm}(\vartheta,\varphi). \tag{3.22}$$

The TE component of the magnetic field is found by substituting the above expression into Maxwell equation (3.20):

$$\mathbf{B}_{\mathrm{TE}}^{\mathrm{quadr}} = -i \sum_{n=1}^{\infty} \sum_{m=-n}^{n} \frac{a_{nm}}{n(n+1)} [\nabla\times(j_n(kr)\hat{\mathbf{L}} Y_{nm}(\vartheta,\varphi))]. \tag{3.23}$$

Let us now consider the TM field components. For this purpose, we first find the radial electric-field component. In that case, there also holds true the important identity

$$(\mathbf{r}\cdot\mathbf{E}_{\mathrm{TM}}^{\mathrm{quadr}}) = e\{\mathbf{r}\cdot[\nabla\times(\nabla\times\delta\mathbf{r}_0)]\}(\mathbf{d}_1\cdot\nabla) \frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|} = e(\mathbf{d}_1\cdot\nabla')\{\delta\mathbf{r}_0\cdot[\nabla'\times(\mathbf{r}'\times\nabla')]\} \frac{e^{ik|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|}. \tag{3.24}$$

Substituting expansion (3.15) into identity (3.24), we obtain the following expression for the radial electric-field component in terms of spherical harmonics:

$$(\mathbf{r}\cdot\mathbf{E}^{\mathrm{quadr}}) = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} b_{nm} j_n(kr) Y_{nm}(\vartheta,\varphi), \tag{3.25}$$

where

$$b_{nm} = -4\pi ke(\mathbf{d}_1\cdot\nabla)[\delta\mathbf{r}_0\cdot(\nabla'\times\hat{\mathbf{L}}')] h_n^{(1)}(kr') \times Y_{nm}^*(\vartheta',\varphi'). \tag{3.26}$$

With the radial electric-field component known, we find the TM field components in exactly the same way as their TE counterparts:

$$\mathbf{B}_{\mathrm{TM}}^{\mathrm{quadr}} = -k \sum_{n=1}^{\infty} \sum_{m=-n}^{n} \frac{b_{nm}}{n(n+1)} h_n^{(1)}(kr) \hat{\mathbf{L}} Y_{nm}(\vartheta,\varphi), \tag{3.27}$$

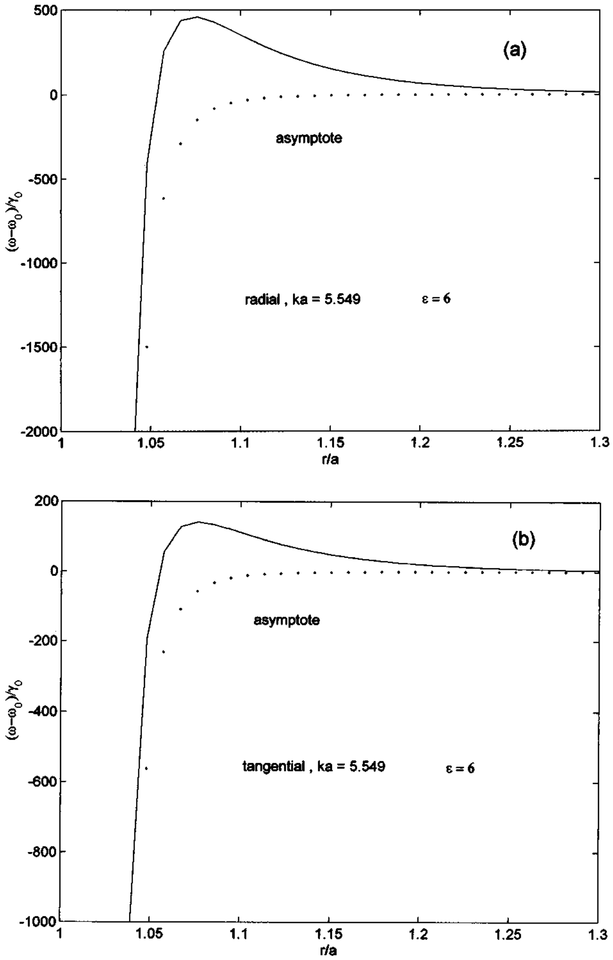


FIG. 6. Frequency shift of quadrupole transitions in the case of resonance interaction with a dielectric microsphere of a sufficiently large radius ($ka=5.5491$) compared to the radiation wavelength as a function of the distance r/a to the surface of the microsphere: (a) radial orientation; (b) tangential orientation. The dashed lines shows long-wave asymptotic relations (4.8) and (4.10)

.

$$\mathbf{E}_{\mathrm{TM}}^{\mathrm{quadr}}=-i\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{b_{nm}}{n(n+1)} \times[\boldsymbol{\nabla}\times(h_n^{(1)}(kr)\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi))]. \tag{3.28}$$

Collecting expressions (3.22), (3.23), (3.27), and (3.28), we obtain the final expansion of the field of the electric dipole in an infinite space in terms of spherical harmonics:

$$\mathbf{B}^{\mathrm{quadr}}=-k\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{b_{nm}}{n(n+1)}h_n^{(1)}(kr)\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi) -i\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{a_{nm}}{n(n+1)} \times\{\boldsymbol{\nabla}\times[h_n^{(1)}(kr)\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi)]\}, \tag{3.29}$$

$$\mathbf{E}^{\mathrm{quadr}}=-i\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{b_{nm}}{n(n+1)} \times\{\boldsymbol{\nabla}\times(h_n^{(1)}(kr)\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi)]\} +k\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{a_{nm}}{n(n+1)}h_n^{(1)}(kr)\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi). \tag{3.30}$$

The total field is the sum of the quadrupole field in free space and the reflected field of interest to us, the expansion of which in terms of spherical harmonics outside the sphere, may be represented in the following form:

$$\mathbf{B}^{(1)}=-k\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{\alpha_{nm}^{(1)}}{n(n+1)}h_n^{(1)}(kr)\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi) -i\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{\beta_{nm}^{(1)}}{n(n+1)}\{\boldsymbol{\nabla}\times[h_n^{(1)}(kr)\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi)]\}, \tag{3.31}$$

$$\mathbf{E}^{(1)}=-i\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{\alpha_{nm}^{(1)}}{n(n+1)}\{\boldsymbol{\nabla}\times[h_n^{(1)}(kr)\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi)]\} +k\sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{\beta_{nm}^{(1)}}{n(n+1)}h_n^{(1)}(kr)\hat{\mathbf{L}}Y_{nm}(\vartheta,\varphi). \tag{3.32}$$

Similar expressions hold inside the sphere, too, if the spherical Hankel functions of the first kind are replaced by spherical Bessel functions.

Now, using the continuity equation for the tangential components of the total field on the surface of the sphere, we can find the unknown coefficients $\alpha_{nm}^{(1)}$ and $\beta_{nm}^{(1)}$, the expressions for which assume the form

$$\alpha_{nm}^{(1)}=\frac{\{\varepsilon_1[z_2h_n^{(1)}(z_2)]'h_n^{(1)}(z_1)-\varepsilon_2[z_1h_n^{(1)}(z_1)]'h_n^{(1)}(z_2)\}}{\{\varepsilon_2[z_1j_n(z_1)]'h_n^{(1)}(z_2)-\varepsilon_1[z_2h_n^{(1)}(z_2)]'j_n(z_1)\}} \times b_{nm}=q_nb_{nm}, \tag{3.33}$$

$$\beta_{nm}^{(1)}=\frac{\{[z_2h_n^{(1)}(z_2)]'h_n^{(1)}(z_1)-[z_1h_n^{(1)}(z_1)]'h_n^{(1)}(z_2)\}}{\{[z_1j_n(z_1)]'h_n^{(1)}(z_2)-[z_2h_n^{(1)}(z_2)]'j_n(z_1)\}}a_{nm} =p_na_{nm}, \tag{3.34}$$

where use is made of the notation $z_{1,2}=k_{1,2}a$, and the coefficients p_n and q_n coincide with the like coefficients (2.3). Thus, the approach suggested has enabled us completely to define, by means of simple calculations, the field of an electric dipole located anywhere outside a dielectric sphere. To find the line parameters in accordance with (3.4) and (3.5), it is necessary to calculate the expression

$$(\mathbf{d}_1\cdot\boldsymbol{\nabla})(\delta\mathbf{r}_0\cdot\mathbf{E}^{(1)}). \tag{3.35}$$

Using formulas (3.31) and (3.32), one can easily reduce the above expression to the form

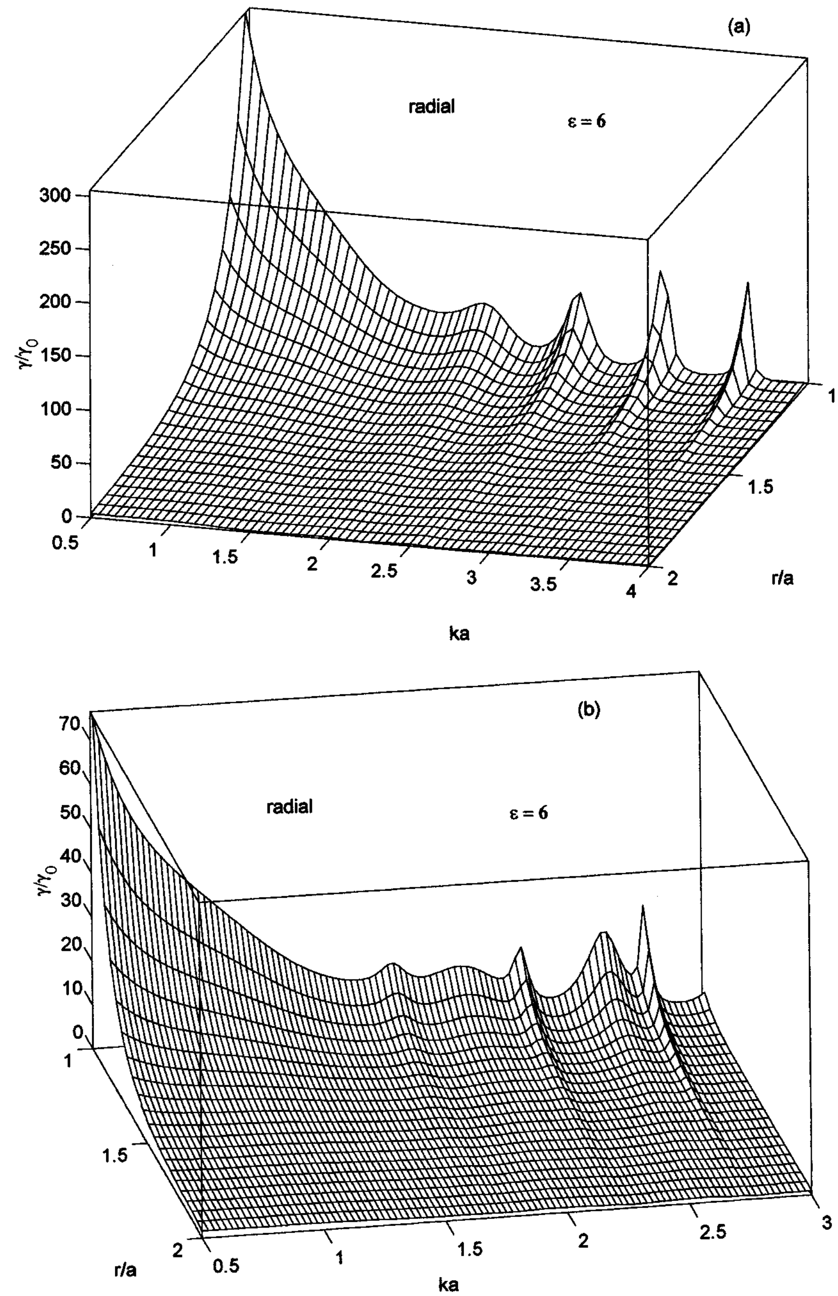


FIG. 7. Linewidth of quadrupole transitions in the neighborhood of a microsphere with $\varepsilon=6$ (diamond) as a function of the distance r/a to the surface of the microsphere and its radius ka: (a) radial orientation; (b) tangential orientation.

$$(\mathbf{d}_1\cdot\boldsymbol{\nabla})(\delta\mathbf{r}_0\cdot\mathbf{E}^{(1)})|_{\mathbf{r}=\mathbf{r}'}=-\frac{i}{e}4\pi k\left\{\sum_{n=1}^{\infty}\sum_{m=-n}^{n}q_n\frac{\widetilde{b}^*_{nm}b^*_{nm}}{n(n+1)}+\sum_{n=1}^{\infty}\sum_{m=-n}^{n}p_n\frac{\widetilde{a}^*_{nm}a^*_{nm}}{n(n+1)}\right\},\tag{3.36}$$

where

$$\begin{aligned}
b^*_{nm}&=e(\mathbf{d}_1\cdot\nabla')\{\delta\mathbf{r}_0[\nabla'\times(h_n^{(1)}(kr')\hat{\mathbf{L}}'Y^*_{nm}(\vartheta',\varphi')]\},\\
\widetilde{b}^*_{nm}&=e(\mathbf{d}_1\cdot\nabla')\{\delta\mathbf{r}_0[\nabla\times'(h_n^{(1)}(kr')\hat{\mathbf{L}}'Y_{nm}(\vartheta',\varphi')]\},\\
a^*_{nm}&=ek(\mathbf{d}_1\cdot\boldsymbol{\nabla}')(\delta\mathbf{r}_0\cdot\hat{\mathbf{L}}')Y^*_{nm}(\vartheta',\varphi')h_n^{(1)}(kr'),\\
\widetilde{a}^*_{nm}&=ek(\mathbf{d}_1\cdot\boldsymbol{\nabla}')(\delta\mathbf{r}_0\cdot\hat{\mathbf{L}}')Y_{nm}(\vartheta',\varphi')h_n^{(1)}(kr').
\end{aligned}\tag{3.37}$$

These expressions are easy to calculate for constant $\mathbf{d}_1$ and $\delta\mathbf{r}_0$ by means of the covariant derivative technique [8].

Substituting expressions (3.36) and (3.37) into (3.7) and (3.8), we obtain the final expressions for the radiation linewidth and frequency shift of a quadrupole of arbitrary orientation:

$$\frac{\gamma}{\gamma_0}=1+\frac{30\pi}{k^4e^2\delta r_0^2d_1^2}\,\mathrm{Re}\left\{\sum_{n=1}^{\infty}\sum_{m=-n}^{n}q_n\frac{\widetilde{b}^*_{nm}b^*_{nm}}{n(n+1)}+\sum_{n=1}^{\infty}\sum_{m=-n}^{n}p_n\frac{\widetilde{a}^*_{nm}a^*_{nm}}{n(n+1)}\right\},\tag{3.38}$$

$$\frac{\Delta\omega}{\gamma_0}=\frac{15\pi}{k^4e^2\delta r_0^2d_1^2}\,\mathrm{Im}\left\{\sum_{n=1}^{\infty}\sum_{m=-n}^{n}q_n\frac{\widetilde{b}^*_{nm}b^*_{nm}}{n(n+1)}+\sum_{n=1}^{\infty}\sum_{m=-n}^{n}p_n\frac{\widetilde{a}^*_{nm}a^*_{nm}}{n(n+1)}\right\}.\tag{3.39}$$

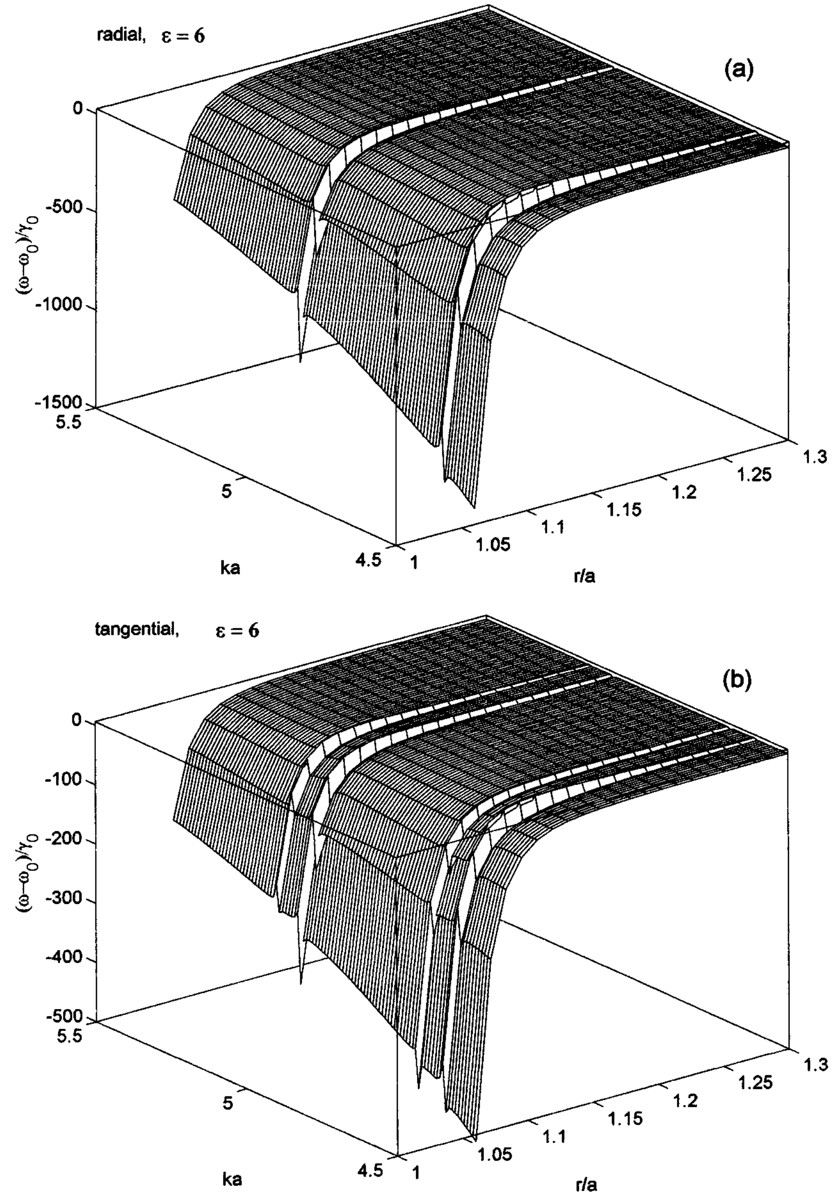


FIG. 8. Frequency shift of quadrupole transitions in the vicinity of a microsphere with $\varepsilon=6$ (diamond) as a function of the distance r/a to the surface of the microsphere and its radius ka: (a) radial orientation; (b) tangential orientation.

Now consider the case where the axis of the dipole $\mathbf{d}_1$ and the oscillation direction $\delta\mathbf{r}_0$ are radially oriented. In that case, it is only the TM modes that are excited; i.e., it is only the coefficients b that are other than zero:

$$b_{nm}^{*}=ie\,\delta r_0 d_1 n(n+1)Y_{nm}^{*}\,\frac{\partial}{\partial r}\left(\frac{h_n^{(1)}(kr)}{r}\right),$$
$$\widetilde{b}_{nm}^{*}=ie\,\delta r_0 d_1 n(n+1)Y_{nm}\,\frac{\partial}{\partial r}\left(\frac{h_n^{(1)}(kr)}{r}\right). \tag{3.40}$$

Here and elsewhere we shall omit primes in atom coordinate.

Substituting (3.40) into (3.38) and (3.39), and using the well-known relation

$$\sum_m Y_{nm}Y_{nm}^{*}=\frac{2n+1}{4\pi}$$

we obtain expressions for the variation of the radiation line parameters of a radially oriented quadrupole in the vicinity of a dielectric sphere ($z=kr$):

$$\frac{\gamma_{\mathrm{rad}}^{Q}}{\gamma_{\mathrm{vac}}^{Q}}=1-\frac{15}{2}\sum_{n=1}^{\infty}n(n+1)(2n+1)\mathrm{Re}\;q_n\left[\frac{d}{dz}\left(\frac{h_n^{(1)}(z)}{z}\right)\right]^2, \tag{3.41}$$

$$\frac{(\omega-\omega_0)_{\mathrm{rad}}^{Q}}{\gamma_{\mathrm{vac}}^{Q}}=-\frac{15}{4}\sum_{n=1}^{\infty}n(n+1)(2n+1)\times\mathrm{Im}\;q_n\left[\frac{d}{dz}\left(\frac{h_n^{(1)}(z)}{z}\right)\right]^2. \tag{3.42}$$

Let us now consider the case where the axis of the dipole $\mathbf{d}_1$ and the oscillation direction $\delta\mathbf{r}_0$ are oriented tangentially (along the φ or θ axis). In that case, both the TM and TE modes are excited i.e., all the coefficients are nonzero:

$$a^*_{nm}=iekd_1\delta r_0\,\frac{h_n^{(1)}(kr)}{r}\,\frac{\partial}{\partial\vartheta}\,\frac{1}{\sin\,\vartheta}\,\frac{\partial Y^*_{nm}}{\partial\varphi},$$

$$\tilde{a}^*_{nm}=iekd_1\delta r_0\,\frac{h_n^{(1)}(kr)}{r}\,\frac{\partial}{\partial\vartheta}\,\frac{1}{\sin\,\vartheta}\,\frac{\partial Y_{nm}}{\partial\varphi}, \tag{3.43}$$

$$b^*_{nm}=ie\,\delta r_0 d_1\left\{\frac{h_n^{(1)}(kr)}{r^2}\,n(n+1)Y^*_{nm}+\frac{1}{r^2}\,\frac{\partial}{\partial r}\,(rh_n^{(1)}(kr))\,\frac{\partial^2 Y^*_{nm}}{\partial\vartheta^2}\right\},$$

$$\tilde{b}^*_{nm}=ie\,\delta r_0 d_1\left\{\frac{h_n^{(1)}(kr)}{r^2}\,n(n+1)Y_{nm}+\frac{1}{r^2}\,\frac{\partial}{\partial r}\,(rh_n^{(1)}(kr))\,\frac{\partial^2 Y_{nm}}{\partial\vartheta^2}\right\}.$$

Substituting (3.43) into (3.38) and (3.39), we obtain the expressions for the variation of the radiation line parameters of a tangentially oriented quadrupole in the presence of a dielectric sphere ($z=kr$):

$$\begin{aligned}\frac{\gamma^Q_{\tan}}{\gamma^Q_{\rm vac}}=1&-\frac{15}{8}\sum_{n=1}^{\infty}n(n+1)(2n+1)\mathrm{Re}q_n\left[\frac{d}{dz}\left(\frac{h_n^{(1)}(z)}{z}\right)\right]^2\\&-\frac{15}{16}\sum_{n=1}^{\infty}(n-1)(n+2)(2n+1)\\&\times\mathrm{Re}q_n\left[\frac{1}{z^2}\,\frac{d}{dz}\,(zh_n^{(1)}(z))\right]^2-\frac{15}{16}\sum_{n=1}^{\infty}(n-1)(n+2)\\&\times(2n+1)\mathrm{Re}\,p_n\left[\frac{1}{z}\,(h_n^{(1)}(z))\right]^2,\end{aligned} \tag{3.44}$$

$$\begin{aligned}\frac{(\omega-\omega_0)^Q_{\tan}}{\gamma^Q_{\rm vac}}=&-\frac{15}{16}\sum_{n=1}^{\infty}n(n+1)(2n+1)\\&\times\mathrm{Im}q_n\left[\frac{d}{dz}\left(\frac{h_n^{(1)}(z)}{z}\right)\right]^2\\&-\frac{15}{32}\sum_{n=1}^{\infty}(n-1)(n+2)(2n+1)\\&\times\mathrm{Im}q_n\left[\frac{1}{z^2}\,\frac{d}{dz}\,(zh_n^{(1)}(z))\right]^2\\&-\frac{15}{32}\sum_{n=1}^{\infty}(n-1)(n+2)(2n+1)\\&\times\mathrm{Im}p_n\left[\frac{1}{z}\,(h_n^{(1)}(z))\right]^2.\end{aligned} \tag{3.45}$$

So in this section we have obtained explicit expressions for the linewidth variation and frequency shift of a classical quadrupole in the neighborhood of a dielectric sphere.

IV. DISCUSSION OF THE RESULTS

In the preceding sections, expressions have been obtained for the variation of the line parameters in the classical and quantum-mechanical approaches. In the present section, we will demonstrate the complete equivalence of these descriptions as applied to the linewidth, and numerically analyze the expressions obtained.

To prove the equivalence of the expressions, use will be made of the easy-to-prove identity

$$|p-\tfrac{1}{2}(1+e^{i\psi})(p+iq)|^2=p^2-\mathrm{Re}[\tfrac{1}{2}(1+e^{i\psi})(p+iq)^2]. \tag{4.1}$$

In our case, the coefficients p and q may be represented in the form

$$\begin{aligned}p_n&=\tfrac{1}{2}(1+e^{i\psi_1}),\\q_n&=\tfrac{1}{2}(1+e^{i\psi_2}),\end{aligned} \tag{4.2}$$

and so the quantum-mechanical expressions (2.19) and (2.24) for the relative linewidth may be represented, by means of (4.1), in the form

$$\frac{\gamma^Q_{\rm rad}}{\gamma^Q_{\rm vac}}=\frac{15}{2}\sum_{n=1}^{\infty}n(n+1)(2n+1)\left\{\frac{d}{dz}\left(\frac{j_n(z)}{z}\right)\right\}^2-\frac{15}{2}\sum_{n=1}^{\infty}n(n+1)(2n+1)\mathrm{Re}\left[q_n\left\{\frac{d}{dz}\left(\frac{h_n^{(1)}(z)}{z}\right)\right\}^2\right], \tag{4.3}$$

$$\begin{aligned}\frac{\gamma^Q_{\tan}}{\gamma^Q_{\rm vac}}=&\frac{15}{8}\sum_{n=1}^{\infty}n(n+1)(2n+1)\left\{\frac{d}{dz}\left(\frac{j_n(z)}{z}\right)\right\}^2+\frac{15}{16}\sum_{n=1}^{\infty}(2n+1)(n-1)(n+2)\left[\frac{1}{z^2}\,\frac{d}{dz}\,(zj_n(z))\right]^2\\&+\frac{15}{16}\sum_{n=1}^{\infty}(2n+1)(n-1)(n+2)\left[\frac{1}{z}\,(j_n(z))\right]^2-\frac{15}{8}\sum_{n=1}^{\infty}n(n+1)(2n+1)\mathrm{Re}q_n\left[\frac{d}{dz}\left(\frac{h_n^{(1)}(z)}{z}\right)\right]^2\\&-\frac{15}{16}\sum_{n=1}^{\infty}(2n+1)(n-1)(n+2)\mathrm{Re}q_n\left[\frac{1}{z^2}\,\frac{d}{dz}\,(zh_n^{(1)}(z))\right]^2-\frac{15}{16}\sum_{n=1}^{\infty}(2n+1)(n-1)(n+2)\mathrm{Re}p_n\left[\frac{1}{z}\,(h_n^{(1)}(z))\right]^2.\end{aligned} \tag{4.4}$$

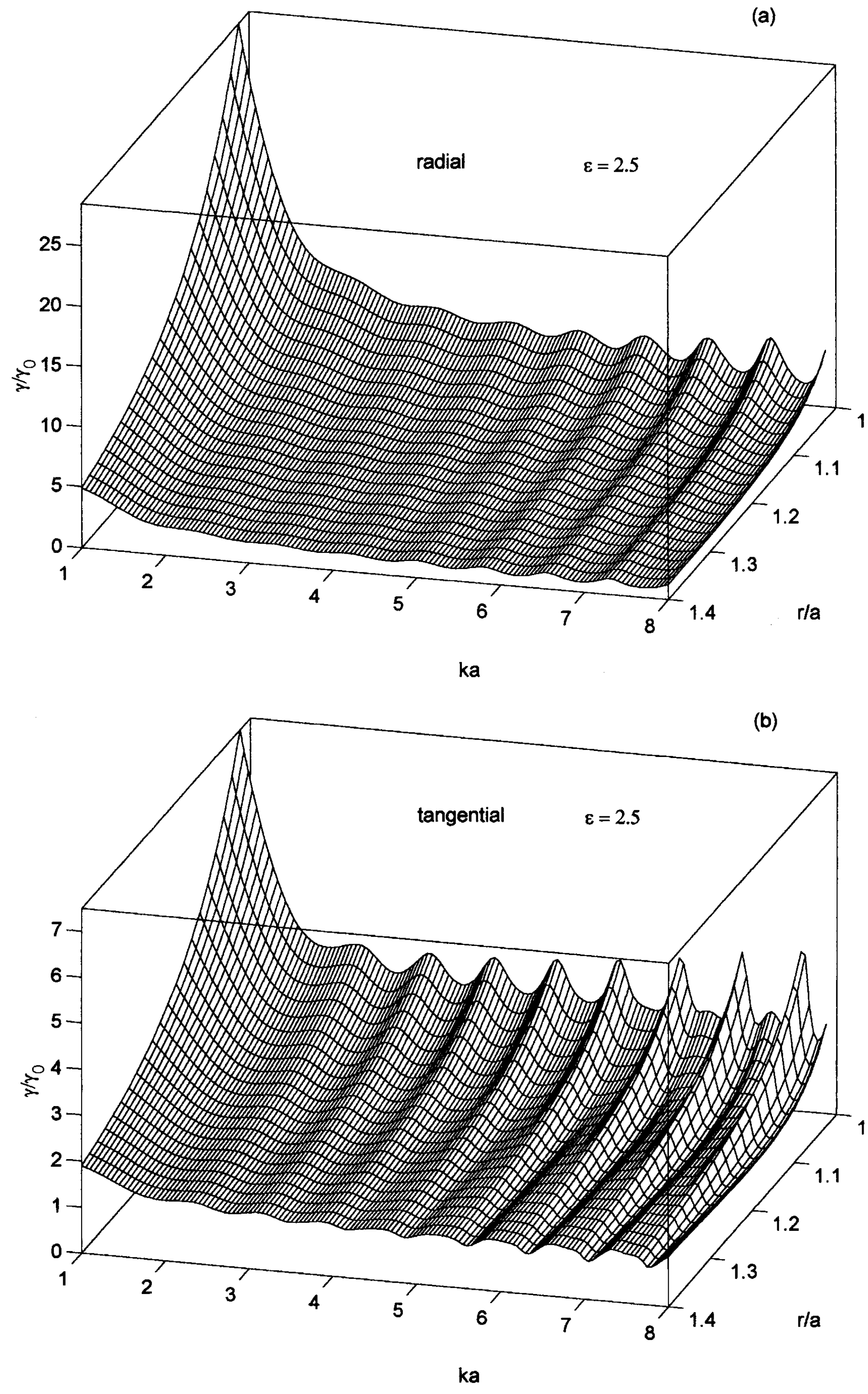


FIG. 9. Linewidth of quadrupole transitions in the neighborhood of a microsphere with $\varepsilon=2.5$ (glass) as a function of the distance r/a to the surface of the microsphere and its radius ka: (a) radial orientation); (b) tangential orientation.

Using the identities

$$\tfrac{2}{15}=\sum_{n=1}^{\infty} n(n+1)(2n+1)\left\{\frac{d}{dz}\left(\frac{j_n(z)}{z}\right)\right\}^2, \qquad (4.5)$$

$$\tfrac{4}{5}=\sum_{n=1}^{\infty}(2n+1)(n-1)(n+2)\left[\frac{1}{z^2}\frac{d}{dz}\,(j_n(z))\right]^2 + \sum_{n=1}^{\infty}(2n+1)(n-1)(n+2)\left[\frac{1}{z}\,(j_n(z))\right]^2, \qquad (4.6)$$

we reduce expressions (4.3) and (4.4) to expressions (3.41) and (3.44), which proves the equivalence of the quantum-mechanical and classical approaches in the calculation of the linewidth of quadrupole radiation. Of course, this equivalence does not relate to frequency shifts, for even in the case of dipole the quantum-mechanical frequency shift results contain such terms as are altogether absent in the classical expressions [10].

Let us consider the expressions found in greater detail. Especially interesting seems to be the case where the atom is located close enough to the surface of a sphere. In the radial

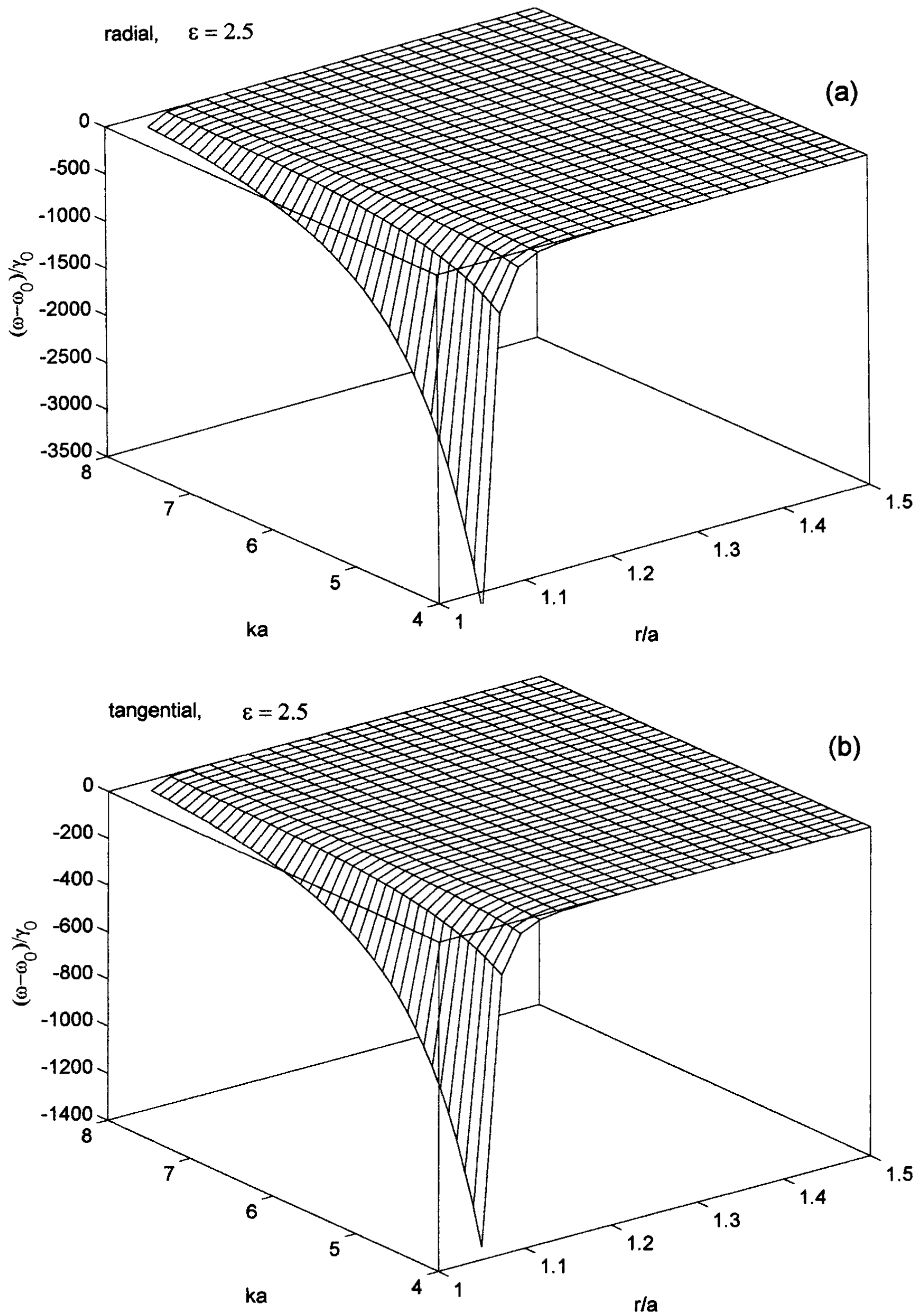


FIG. 10. Frequency shift of quadrupole transitions in the neighborhood of a microsphere with $\varepsilon=2.5$ (glass) as a function of the distance r/a to the surface of the microsphere and its radius ka: (a) radial orientation; (b) tangential orientation.

case of an atom located close to the surface of a sphere of a small radius ($ka \to 0$), expressions (3.41) and (3.42) for the relative linewidth and relative frequency shift assume the form

$$\frac{\gamma_{\rm rad}^{Q}}{\gamma_{\rm vac}^{Q}}=\frac{180}{(ka)^2}\left[\frac{\varepsilon-1}{\varepsilon+2}\right]^2+\cdots\,, \tag{4.7}$$

$$\frac{(\omega-\omega_0)_{\rm rad}^{Q}}{\gamma_{\rm vac}^{Q}}=-\frac{45}{16}\,\frac{\varepsilon-1}{\varepsilon+1}\,\frac{1}{(z-z_2)^5}, \tag{4.8}$$

where $z=rk$, $z_2=ak$, and $k=\omega/c$.

In the case of tangential orientation of the principal axis of the quadrupole, expressions (3.44) and (3.45) for the relative linewidth and relative frequency shift assume the form

$$\frac{\gamma_{\rm tan}^{Q}}{\gamma_{\rm vac}^{Q}}=\frac{45}{(ka)^2}\left[\frac{\varepsilon-1}{\varepsilon+2}\right]^2+\cdots\,, \tag{4.9}$$

$$\left(\frac{\omega-\omega_0}{\gamma_{\rm vac}^{Q}}\right)^{\rm rad}=-\frac{135}{128}\,\frac{\varepsilon-1}{\varepsilon+1}\,\frac{1}{(z-z_2)^5}. \tag{4.10}$$

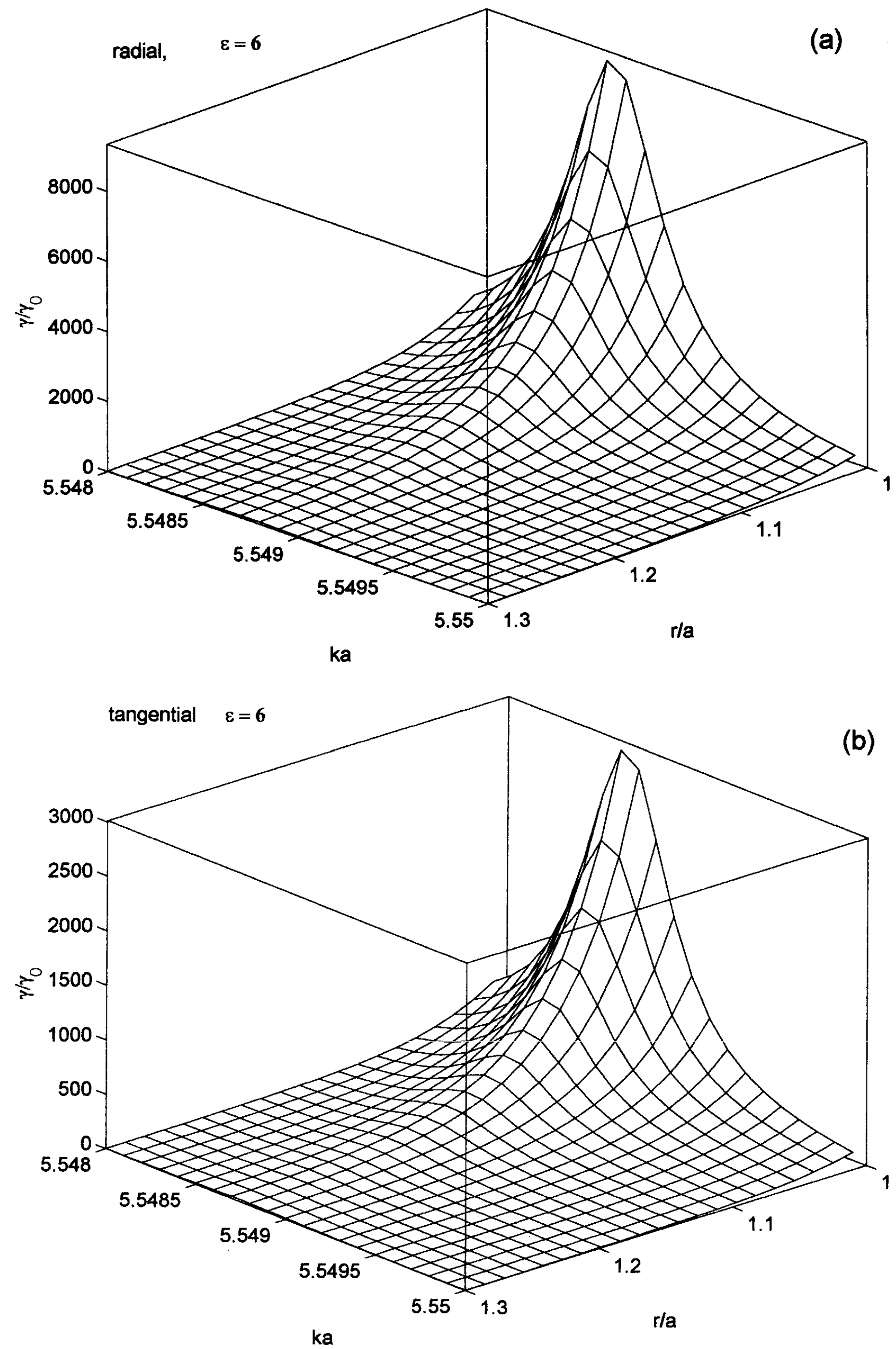


FIG. 11. Linewidth of quadrupole transitions in close proximity to a microsphere with $\varepsilon=6$ (diamond) as a function of the distance r/a to the surface of the microsphere and its radius ka in the vicinity of a TM resonance ($ka=5.5491$): (a) radial orientation; (b) tangential orientation.

It is of interest to compare the results obtained with expressions for the linewidth and frequency shift in the case of dipole transitions, which in our notation have the form [11]

$$\left(\frac{\gamma}{\gamma_0}\right)_{\mathrm{dip}}=1-\tfrac{3}{2}\ \mathrm{Re}\left[\cos^2\psi\sum_{n=1}^{\infty} n(n+1)(2n+1)q_n\left(\frac{h_n^{(1)}(z)}{z}\right)^2+\sin^2\psi\sum_{n=1}^{\infty}(n+1/2)\left\{p_n(h_n^{(1)}(z))^2+q_n\left(\frac{d(zh_n^{(1)}(z))}{zdz}\right)^2\right\}\right], \tag{4.11}$$

$$\left(\frac{\omega-\omega_0}{\gamma_0}\right)_{\mathrm{dip}}=-\tfrac{3}{4}\ \mathrm{Im}\left[\cos^2\psi\sum_{n=1}^{\infty} n(n+1)(2n+1)q_n\left(\frac{h_n^{(1)}(z)}{z}\right)^2+\sin^2\psi\sum_{n=1}^{\infty}(n+\tfrac{1}{2})\left\{p_n(h_n^{(1)}(z))^2+q_n\left(\frac{d(zh_n^{(1)}(z))}{zdz}\right)^2\right\}\right], \tag{4.12}$$

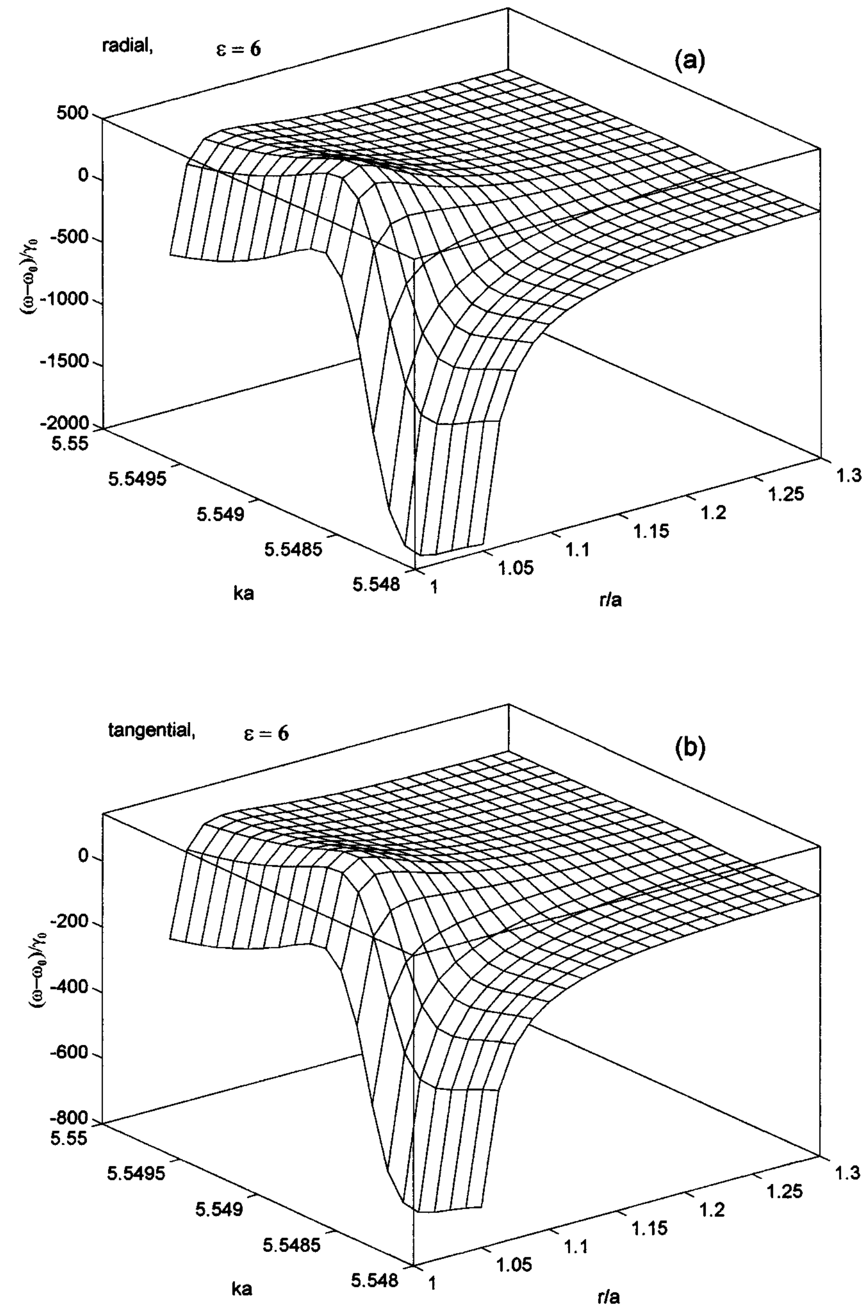


FIG. 12. Frequency shift of quadrupole transitions in close proximity to a microsphere with $\varepsilon=6$ (diamond) as a function of the distance r/a to the surface of the microsphere and its radius ka in the vicinity of a TM resonance ($ka=5.5491$): (a) radial orientation; (b) tangential orientation.

where ψ is the angle between the dipole axis and the radius.

At small distances from the surface of a small sphere, we have, instead of expressions (4.11) and (4.12), the following asymptotic expressions:

$$\frac{\gamma_{\text{rad}}^{\text{dip}}}{\gamma_{\text{vac}}^{\text{dip}}}=\left[\frac{3\varepsilon}{\varepsilon+2}\right]^2+\cdots , \qquad (4.13)$$

$$\frac{\gamma_{\tan}^{\text{dip}}}{\gamma_{\text{vac}}^{\text{dip}}}=\left[\frac{3}{\varepsilon+2}\right]^2+\cdots \qquad (4.14)$$

and

$$\left(\frac{\omega-\omega_0}{\gamma_{\text{vac}}^{\text{dip}}}\right)^{\text{rad}}=-\frac{3}{16}\frac{\varepsilon-1}{\varepsilon+1}\frac{1}{(z-z_2)^3}, \qquad (4.15)$$

$$\left(\frac{\omega-\omega_0}{\gamma_{\text{vac}}^{\text{dip}}}\right)^{\tan}=-\frac{3}{32}\frac{\varepsilon-1}{\varepsilon+1}\frac{1}{(z-z_2)^3}. \qquad (4.16)$$

Comparing between (4.7) and (4.9) and (4.13) and (4.14), it is not very difficult to establish the fact of substantial acceleration of quadrupole transitions. Moreover, one can see from these expressions that as the size of the microsphere is reduced in comparison with the radiation wavelength, the quadrupole transition rate can increase beyond all bounds. Of course, one should verify the applicability of the perturbation theory here.

Figure 3 shows the relative radiation linewidth for quadrupole and dipole transitions in atoms located near to the surface of a sphere as a function of the sphere radius ka in the case of radial orientation, whence the substantial acceleration of the quadrupole transitions as compared to their dipole counterparts is clearly evident. The crosses in the figure indicate asymptotic relations (4.7) and (4.9).

Comparison between (4.8) and (4.10) and (4.15) and (4.16) shows a substantial increase in the frequency shift of the quadrupole transition in comparison with the dipole transition as one draws closer to the surface of the sphere, the index of singularity changing from 3 to 5.

As already noted, these facts are due to the quadrupole sensitivity to the field gradient which is singular in close proximity to the surface of a small-radius sphere. Figure 4 presents the linewidths of quadrupole and dipole transitions as a function of the distance r/a to the surface of the microsphere for various orientations and various refractive indices of the microsphere [$ka=0.01$, $\varepsilon=6$ (diamond), and $\varepsilon=2.5$ (glass)]. One can see from the figure that a substantial increase in the quadrupole transition rate occurs in the case of both glass and diamond microsphere.

Shown in Fig. 5 is the linewidth of quadrupole and dipole transitions for various orientations in the case of resonance interaction with a dielectric sphere of sufficiently great radius ($ka=5.5491$) in comparison with the radiation wavelength as a function of the distance r/a to the surface of the sphere. It is evident from the figure that even in that case there takes place an acceleration of the quadrupole transitions. Figure 6 illustrates expressions for the frequency shift in the neighborhood of this resonance, as well as the corresponding asymptotic relations (4.8) and (4.10).

Figures 7 and 8 present the linewidth and frequency shift of quadrupole transitions in the vicinity of a diamond microsphere as a function of the distance r/a to the surface of the microsphere and its radius ka. Figures 9 and 10 show similar curves for a glass microsphere. Clearly seen in these figures are the resonance interaction regions, the greater number of resonances in the case of tangential orientation of the quadrupole axis being due to the emergence of TM modes.

Presented in Figs. 11 and 12 are the linewidth and frequency shift of quadrupole transitions in close proximity to a diamond microsphere as a function of the distance r/a to its surface and its radius ka in the neighborhood of a TM resonance ($ka=5.5491$). These curves are of distinct dispersion character.

Thus in the present work we have considered quadrupole radiation processes in the neighborhood of a dielectric microsphere. We have demonstrated by way of explicit calculations that the classical and quantum-mechanical approaches yield the same expressions for the quadrupole transition linewidth. We have also found a classical expression for the frequency shift of quadrupole transitions. The analysis of the expressions found has shown that quadrupole transitions are much more sensitive to changes in the structure of electromagnetic field associated with the presence of the dielectric microsphere. The results obtained are easy to generalize to the case of higher-order multipoles, it being evident within the framework of the approach suggested that the sensitivity of the atom to the presence of a microsphere will be even higher in that case.

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