

# Effect of surface deformations of spherical microparticles on Q-factor of their resonance modes: geometric optics approach

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The effect of small surface deformations of spherical microparticles on the Q-factor of their natural resonance modes has been studied in detail. Based on the geometric optics representation of a natural mode of a sphere as a congruence of optical rays confined between the particle surface and the internal caustic, a simple equation is derived that relates the Q-factor of a mode with the surface deformation amplitude. The values of the Q-factor of deformed water droplets are compared with similar results obtained by the perturbation method of the wave theory.

Micron-size spherical particles in the optical wavelength region can be considered as open cavities having a set of high-Q resonance modes. Theoretically, the Q-factor of some of these modes often referred to as whispering gallery (WG) modes can achieve rather high values.<sup>1</sup> However, the Q-factor higher than  $\sim 10^8$  has not been observed in practice yet.<sup>2</sup> One of the possible causes for this is a non-spherical shape of particles. As to solid particles, their spherical shape can be distorted by inhomogeneities formed in the process of their generation. The shape of liquid aerosol particles is subject to surface deformations due to both natural causes (temperature fluctuations, air flows) and the ponderomotive forces due to the action of an intense optical field.<sup>3</sup>

The Q-factor of a particle-microcavity is a very important characteristic affecting the processes of nonlinear interaction of light fields in such systems.<sup>4</sup> At the same time, the Q-factor itself depends not only on the optical characteristics of a substance, but also on the geometric shape of a microcavity. As to spherical particles, any deformations of their surface finally lead to deterioration of resonance properties. The most significant cause of the decrease of the Q-factor is perturbation of phase matching conditions for resonance of optical modes in deformed particles.<sup>5</sup> This changes the spatial structure of natural oscillations and leads to a shift in the frequency position of resonances.

As known, open cavities, among which there are low-absorbing dielectric particles, have an additional channel for dissipation of stored electromagnetic energy, namely, radiative losses. The natural frequencies of electromagnetic oscillations in such systems prove to be significantly complex, and the ratio of their real  $\omega'$  and imaginary  $\omega''$  parts  $Q = \omega' / 2\omega''$  is often used to determine their Q-factor. In view of this circumstance, the problem of studying the resonance characteristics of deformed spherical particles is closely connected with the problem of determining the frequencies of their natural

electromagnetic oscillations, that is, solution of the Helmholtz equation for the electric field vector  $\mathbf{E}(\mathbf{r})$ :

$$\text{rot rot } \mathbf{E}(\mathbf{r}) + \varepsilon_a \frac{\omega^2}{c^2} \mathbf{E}(\mathbf{r}) = 0 \quad (1)$$

under condition of continuity of its tangential components at the perturbed particle surface:

$$[\mathbf{E}_i(\mathbf{r}) - \mathbf{E}_s(\mathbf{r})] \times \mathbf{n}_r = 0, \quad [\mathbf{H}_i(\mathbf{r}) - \mathbf{H}_s(\mathbf{r})] \times \mathbf{n}_r = 0, \quad (2)$$

where  $\varepsilon_a$  is the dielectric constant of the particle substance;  $\mathbf{n}_r$  is the external normal to the particle surface;  $c$  is the speed of light in vacuum, and the subscripts  $i$  and  $s$  stand for the field inside the particle and outside of it, respectively. The solution of Eqs. (1) and (2) for a sphere at  $\varepsilon_a = \text{const}$  is well-known and can be presented in the form of two systems of orthogonal functions specifying the  $\text{TE}_{nm}$  and  $\text{TH}_{nm}$  electromagnetic modes<sup>6</sup>:

$$\begin{aligned} \mathbf{E}_{s_{nm}}(\mathbf{r}) &= A_{nm} \nabla \times [\mathbf{M}_{nm}(\theta, \varphi) \xi_n(kr)] + \\ &+ B_{nm} \mathbf{M}_{nm}(\theta, \varphi) \xi_n(kr), \\ \mathbf{E}_{i_{nm}}(\mathbf{r}) &= C_{nm} \nabla \times [\mathbf{M}_{nm}(\theta, \varphi) \psi_n(\sqrt{\varepsilon_a} kr)] + \\ &+ D_{nm} \mathbf{M}_{nm}(\theta, \varphi) \psi_n(\sqrt{\varepsilon_a} kr). \end{aligned} \quad (3)$$

Here  $A_{nm}$ ,  $B_{nm}$ ,  $C_{nm}$ , and  $D_{nm}$  are the amplitude coefficients;  $\psi_n$  and  $\xi_n$  are the spherical Riccati–Bessel functions;  $\mathbf{M}_{nm}$  are the spherical vector harmonics;  $n$  and  $m$  are the mode indices, whose meaning will be discussed below.

At arbitrary deviations of the particle shape from the ideally spherical, the analytical solution of Eq. (1) can be found only at a small amplitude of deformations. The canonical method for solution of this problem is application of the perturbation theory. This approach was realized in Refs. 7 and 8 and with some modifications in Ref. 9. Lai et al.<sup>7,8</sup> and Datsyuk et al.<sup>9</sup> replaced surface perturbations of a spherical particle with perturbations of the particle dielectric constant at passage

through the particle boundary. As a result, a system of coupled equations with integral coefficients was obtained for the determination of natural frequencies.<sup>7</sup> In the case of symmetric particle deformations described by the function  $f(r, \theta)$ , in the first approximation with respect to the small parameter of the deformation amplitude  $\xi_A$ , this system has the following form:

$$\frac{\Delta k_{nm}}{k_{nm}} = -\frac{V_n^{mm}}{2G_n}, \quad V_n^{mm} = \int_V dr' \xi_A f(r'; \theta) |\mathbf{M}_{nm}|^2,$$

$$G_n = (|\varepsilon_a| - 1) \frac{a_0}{2k^2} |\xi_n(x_a)|^2,$$

where  $k_{nm} = \omega_{nm}/c$ ;  $\Delta k_{nm}$  is the change in the complex wavenumber for the natural mode  $\mathbf{E}_{nm}$  at deformation of the particle with the radius  $a_0$  and the diffraction parameter  $x_a = 2\pi a_0/\lambda$ . The Q-factor of the particle in this case can be represented as a series over the deformation amplitude  $\xi_A$  (Ref. 8):

$$1/Q = 1/Q_0 + C_1 \xi_A + C_2 \xi_A^2 + \dots, \quad (4)$$

where  $Q_0$  is the Q-factor of the ideal sphere, and the coefficients  $C_1$  and  $C_2$  depend on the function  $f(r, \theta)$  and can be expressed through the angular integrals and the combination of logarithmic derivatives of spherical functions.

In Refs. 10–12, to find natural frequencies of oscillations, the perturbation was introduced in the boundary conditions (2), rather than in equation (1) itself. In this case, the structure of eigenfunctions  $\mathbf{E}_{nm}$  was chosen similarly to the structure of eigenfunctions of the ideal sphere, and equations at the particle boundary were written with the allowance for particle deformations. Generalization of this approach to the case of arbitrary types of substance inhomogeneity and perturbation of the particle surface is the method of layer-by-layer T-matrices (transformation matrices) used, for example, in Refs. 13 and 14.

The main disadvantage of the techniques considered above is their complicated numerical realization that requires high-performance computers and voluminous computations. In our opinion, this prevents the use of the results of these theoretical investigations for interpretation of experiments. The proposed method for estimation of the Q-factor of weakly deformed spherical particles is free of this disadvantage and is based on the ray approximation of eigenfunctions of Eq. (1) that was proposed in Ref. 15.

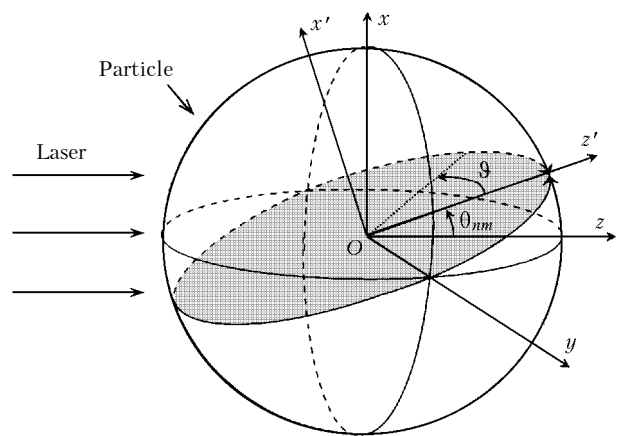
Our approach is based on the assumption that the decrease in Q-factor of a deformed sphere is mostly caused by the shift of the resonance frequency of natural modes from the initial (unperturbed) value. As a consequence, the Q-factor of a particle measured at a fixed frequency of the natural mode decreases, because the resonance conditions for this frequency are distorted. It is obvious that the rate of the decrease of Q-factor in this case is largely determined by the shape of the spectral resonance curve, which, as is well known,<sup>1,4</sup> is described by the Lorentz profile:

$$Q(x)/Q_0 = 1/[1 + (\Delta x/\Gamma)^2], \quad \Delta x = x - x_a, \quad (5)$$

where  $\Gamma = x_a/Q_0$  is the resonance halfwidth;  $x = ka$ . Thus, the main task of this analysis is to correlate particle deformations with variations of the diffraction parameter  $\Delta x$ . The definition of the diffraction parameter as a characteristic of optical properties of, first of all, a spherical body points to the necessity of representing the deformed particle also as a sphere, but with other radius, which is determined by the character and the amplitude of deformations.

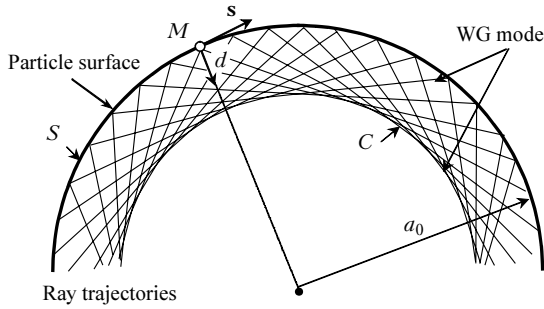
For further consideration of the details of the proposed approach, it is necessary to pass from the wave description to the geometric optics description of the resonance oscillation modes of the dielectric spheres.

As known, the effect of resonance excitation of the optical field inside a spherical particle is observed as the frequency of the pump radiation coincides with the frequency of some WG mode of a dielectric sphere. A WG mode can be presented as a standing wave formed by superposition of two waves propagating in the counter directions. Reflecting from the liquid/ambient medium interface, these waves make a full turn (or several turns) around the droplet surface and come to the initial point in phase, thus forming the positive feedback. The spatial structure of such modes is usually characterized by three integer indices: two angular numbers (mode number  $n$  and azimuth index  $m$ ) and one radial number (mode order  $j$ ). The electromagnetic field of the WG mode has a sharply inhomogeneous spatial profile and concentrates in both the radial direction near the particle surface and the latitudinal direction (at  $n \gg 1$ ). This field occupies an orbit passing through the particle center and lying at the angle  $\theta_{nm} = \arccos(m/n)$  relative to the equatorial cross section (Fig. 1). The latter circumstance is especially important, since it allows us to pass from the 3D pattern to its 2D analog in the orbit plane in the further investigation of the spatial distribution of fields of particle natural modes.



**Fig. 1.** Scheme of localization of WG mode in a spherical particle:  $\theta_{nm}$  is the polar angle of tilt of the mode orbit;  $\varphi$  is the azimuth angle in the orbit plane of WG mode.

Let us use the geometric optic representation of the WG mode as some congruence (system) of optical rays existing in a closed area bounded by the curve  $S$ , which transforms into itself after a finite number of ray reflections from the interface.<sup>15</sup> Such a system of rays turns out to be bounded in space by the caustic  $C$  on one side and by the surface  $S$  on the other side (Fig. 2), as well as it has the property of stability in the first approximation.



**Fig. 2.** Geometric optics representation of WG mode in a spherical particle:  $C$  is internal caustic;  $S$  is the particle surface of radius  $a_0$ ;  $M$  is an arbitrary point on the particle surface;  $s$  is unit vector tangent to the surface;  $d$  is the distance along the normal from the particle surface.

The structure of the electromagnetic field characterizing the WG mode in a sphere of radius  $a_0$  can be written in terms of the Airy function<sup>16</sup> that is the asymptotic representation ( $n > j$ ,  $n \gg 1$ ) of the traditional resolution of the field of natural modes in terms of spherical harmonics (3):

$$E_{nmj} = \frac{\text{Ai}(d/D_{nj} + \alpha_j)}{|\text{Ai}'(\alpha_j)|\sqrt{D_{nj}}} \exp\{ik_{nj} s\},$$

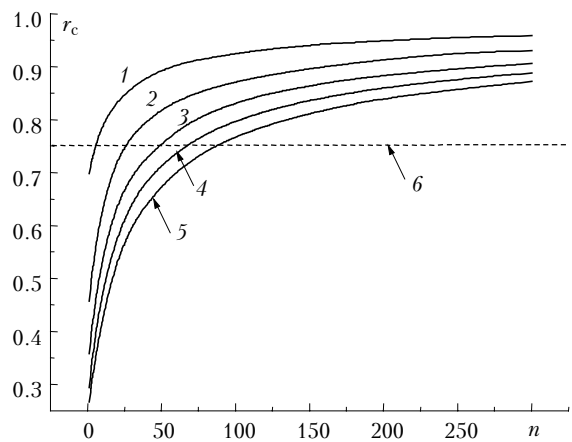
where  $s$  is the arc length of the curve  $S$  measured from some initial point to the point  $M$ ;  $\alpha_j$  is the root of the Airy function ( $\text{Ai}(-\alpha_j) = 0$ );  $k_{nj}$  is the mode eigenvalue. The parameter  $D_{nj} = (2k_{nj}/a_0)^{1/3}$  characterizes the thickness of the layer, the mode field concentrates in, and the equation of caustic  $C$  has the form

$$d = D_{nj} \alpha_j + O[(n/j)^{4/3}].$$

Note, for information, that the grazing angle  $\gamma_{nj}$  of the ray at its reflection from the surface is connected with the mode eigenvalues as follows:

$$\gamma_{nj} = \sqrt{\alpha_{nj}} (9/2a_0 k_{nj})^{1/3}.$$

Figure 3 depicts the dependence of the caustic  $C$  position on the WG mode number  $n$  for several values of its order  $j$ . It can be seen that as the mode number increases, the distance from the caustic to the droplet surface decreases, and, correspondingly, the area occupied by the WG mode decreases. The dependence has the behavior similar with the decrease in the order of the resonance mode.



**Fig. 3.** Dependence of the relative radial coordinate of the caustic  $r_c = (a_0 - d)/a_0$  on the WG mode number  $n$  for different values of the mode order  $j$  (numbers by the curves). The dashed line 6 shows the caustic position for total internal reflection angle.

For a stable system of rays representing the WG mode to be formed, the phase matching conditions should be fulfilled. In terms of geometric optics, these conditions correspond to the so-called conditions of ray phase quantization:

$$k\Lambda = 2\pi n, \quad k\eta = 2\pi(j + 3/4), \quad (6)$$

where  $\Lambda$  is the length of the caustic  $C$ ;  $\eta$  is the sum of lengths of the rays reflected from the surface  $S$  minus the length of the caustic part between tangent points. Solution of the system (6) by the method of successive approximations gives the equation for the eigenvalues  $k_{nj}$  of WG modes in the form of a fractional-power series:

$$k_{nj} = k_{nj}^0 [1 + A_2(k_{nj}^0)^{-2/3} + A_4(k_{nj}^0)^{-4/3} + \dots], \quad n \gg 1. \quad (7)$$

Here  $k_{nj}^0 = 2\pi n/L$ ,  $L$  is the length of the profile of the surface  $S$ , and the coefficients  $A_l$  depend on the curvature of the surface  $S$ :

$$A_2 = \frac{\alpha_j}{2^{1/3} L} \int_0^L \frac{ds}{[\rho(s)]^{2/3}}, \quad A_4 \approx 1/3 A_2^2,$$

where  $\rho(s)$  is the curvature radius of the surface at the point with the coordinate  $s$ .

Deformations of the surface  $S$  obviously change the phase relations in the previous congruence of rays and lead to the situation that the quantization conditions (6) are fulfilled already at somewhat different values of  $k_{nj}$ . From the structure of the series (7), it can be seen that the dependence of the eigenvalues  $k_{nj}$  on the geometry of the reflecting surface is mostly concentrated in the parameter  $L$ . Therefore, if we neglect the dependence of the coefficients  $A_l$  on  $L$  (this is possible, because the curvature radius depends on the shape of the surface  $S$  weaker as compared with the length of surface profile  $L$ ) and denote a small

change of the profile length as  $\delta L$ , then the corresponding shift of eigenvalues  $\delta k_{nj}$  can be written as

$$\frac{\delta k_{nj}}{k_{nj}^0} \approx -\frac{\delta L}{L} [1 + 1/3 A_2(k_{nj}^0)^{-2/3} - 1/3 A_4(k_{nj}^0)^{-4/3} + \dots].$$

It can easily be seen that if the surface profile is a circle, then  $A_2 = \frac{\alpha_j}{2^{1/3} n^{2/3}}$ , and  $A_2 \ll 1$  is valid under the condition  $n \gg 1$ . Consequently, in the “zero” approximation (in terms of  $k_{nj}^0$ ), for  $\delta k_{nj}$  we have

$$\frac{\delta k_{nj}}{k_{nj}^0} = -\frac{\delta L}{L}. \tag{8}$$

Let us set the surface perturbation of a spherical droplet as a function  $\xi_A f_{nm}(\vartheta)$ , where  $\xi_A$  is some perturbation amplitude ( $\xi_A \ll a_0$ ),  $\vartheta$  is the angle in the orbit plane of the  $E_{nmj}$  mode ( $\vartheta \in [0, 2\pi]$ ), and  $f_{nm}(\vartheta)$  is assumed differentiable sufficient number of times with respect to  $\vartheta$  and, besides,  $|f_{nm}| < 1$ . Then the radius of the deformed particle can be written as

$$r(\vartheta) = a_0 [1 + \bar{\xi}_A f_{nm}(\vartheta)], \quad \bar{\xi}_A = \xi_A/a_0. \tag{9}$$

The surface shape set in the form (9) allows us to use the known equation for the differential of an arc of an arbitrary curve<sup>17</sup>:

$$ds = \sqrt{r^2 + (r')^2},$$

where

$$r' = dr/d\vartheta. \tag{10}$$

Substituting Eq. (9) into Eq. (10), we obtain the following equation for  $\delta L$ :

$$\delta L \approx \xi_A \int_0^{2\pi} [f_{nm}(\vartheta) + \bar{\xi}_A/2 \{f_{nm}^2(\vartheta) + [f'_{nm}(\vartheta)]^2\}] d\vartheta,$$

$$f' = df/d\vartheta.$$

On the assumption of small shift of the particle surface  $\bar{\xi}_A$ , we believe that the shape of the deformed profile is also a circle with some effective radius  $a_{ef}$ :

$$a_{ef} = a_0 \left[ 1 + \frac{\delta L}{2\pi a_0} \right] = a_0 [1 + \bar{\xi}_A q_{nm}]. \tag{11}$$

Here  $q_{nm} = \frac{\delta L}{2\pi \xi_A} = q_{nm}^0 + \bar{\xi}_A q_{nm}^1$  is the transformation coefficient. Then the corresponding change of the

diffraction parameter of a new effective sphere with respect to its initial value  $x_a$  can be written as  $(\delta x/x_a) = \bar{\xi}_A q_{nm}$ .

Substitution of Eq. (11) into the equation for the Lorentz profile of the resonance curve (5) gives the sought equation for the Q-factor of the deformed sphere:

$$\frac{1}{Q_D} = \frac{1}{Q_0} + C_2 \bar{\xi}_A^2 + C_3 \bar{\xi}_A^3 + C_4 \bar{\xi}_A^4, \tag{12}$$

where

$$C_2 = Q_0 [q_{nm}^0]^2, \quad C_3 = 2Q_0 q_{nm}^0 q_{nm}^1, \quad C_4 = Q_0 [q_{nm}^1]^2.$$

It should be noted that this equation in its form is similar to the above series (4), which were obtained by the method of the perturbation theory, and it is indicative of the fact that the Q-factor of the deformed sphere for a selected mode can change only toward its decrease.

Let us consider now some particular cases.

### 1. Spheroidal deformations of a particle

The shape of a deformed particle is specified as

$$r(\vartheta) = a_0 [1 + 2e \cos^2(\vartheta)]^{1/2}, \tag{13}$$

where  $e = 1 - [1/(1 + \bar{\xi}_A)]^2$ , and  $a_0$  and  $a_0(1 + \bar{\xi}_A)$  are the lengths of the main spheroid axes. This type of deformations is characteristic of, for example, the initial phase of ponderomotive oscillations of a droplet, when it elongates predominantly along the direction of the radiation-induced force.<sup>3</sup>

Comparing Eqs. (9) and (13), we can find that in this case

$$f_{nm}(\vartheta) \approx \frac{\bar{\xi}_A + 2}{(2\bar{\xi}_A + 1)^2} \cos^2(\vartheta),$$

and the transformation coefficient accurate to the terms on the order of  $\bar{\xi}_A^2$  is equal to

$$q_{nm} \approx 1/4 \frac{\bar{\xi}_A + 2}{(2\bar{\xi}_A + 1)} \approx 1/2, \text{ at } \bar{\xi}_A \ll 1. \tag{14}$$

### 2. Combined (ellipsoidal) deformations

Let us write droplet deformations as

$$r(\vartheta) = a_0 [1 + \bar{\xi}_A \cos(N\vartheta)], \tag{15}$$

where  $N$  is an integer number. Unlike the case considered above, the shape of the deformed surface specified by Eq. (15) rather closely describes natural deformations

of liquid particles due to either their fall in the air or thermal instability of their surface. At  $N = 1$  this type of deformations corresponds to "classical" ellipsoidal deformations of a droplet that are observed at its free oscillations at a fundamental (Rayleigh) frequency.<sup>18</sup>

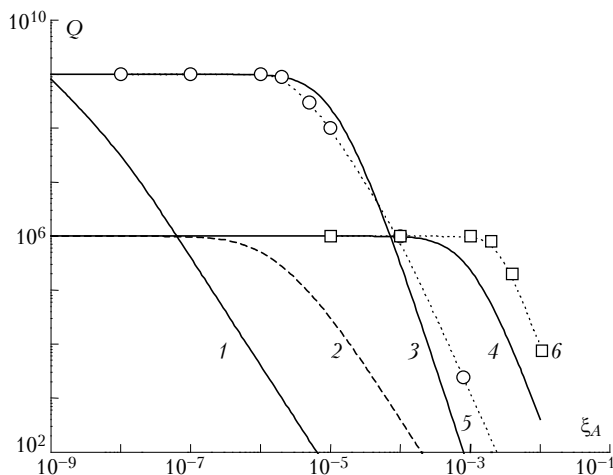
After analogous transformations we obtain

$$f_{nm}(\vartheta) = \cos(N\vartheta),$$

$$q_{nm} = 1/4 \bar{\xi}_A^2 (1 + N^2). \quad (16)$$

It can be seen that the coefficients  $C_2$  and  $C_3$  are zero because of the periodicity of the function  $f_{nm}$  with respect to the angle  $\vartheta$ .

Figure 4 depicts the plot of the Q-factor of two resonance modes  $TE_{97}^1$  and  $TE_{94}^2$  as a function of deformations calculated by Eqs. (14) and (16). For a comparison, this figure also presents the results of numerical calculations of this parameter from Ref. 11. It follows from the figure that, under all other conditions being the same, the spheroidal type of particle deformations leads to a stronger decrease in the Q-factor of resonance modes as compared with the combined ellipsoidal perturbation of the particle surface. The difference in Q-factor values from the results of Ref. 11, especially, for low-Q resonances is a consequence of the used approximation of effective sphere for the Lorentz profile, which gives the higher values of the coefficient  $C_4$  than in the method of small perturbations. However, in general, the results obtained point to the true tendency that particle deformations have the strongest effect just at high-Q resonance modes, whose electromagnetic field concentrates closer to the surface (smaller values of  $D_{nj}$ ).



**Fig. 4.** Q-factors of two WG modes:  $TE_{97}^1$  (curves 1, 3, 5) and  $TE_{94}^2$  (curves 2, 4, 6) vs. the amplitude of surface deformations. Curves 1 and 2 are calculated by Eq. (14); curves 3 and 4 are calculated by Eq. (16); curves 5 and 6 are plotted according to the data of Ref. 11.

Equations (14) and (16) were obtained only for the particular case of the principal particle cross section, ( $\theta_{nm} = 0$ ) corresponding to the WG modes with the azimuth index  $m = \pm n$ . As was noted above, the

electromagnetic field of the modes with  $m \neq n$  is dominantly localized in the planes inclined by the angle  $\theta_{nm} \neq 0$  relative to the principal cross section. Therefore, for them Eqs. (14) and (16) should be corrected.

For this purpose, we can use known equations for transformation of the spatial coordinates.<sup>17</sup> The new coordinate system is connected with the orbit of some natural mode. In fact, it is the initial coordinate system, whose axes  $x'$  and  $z'$  are turned around the axis  $y$  by the angle  $\theta_{nm}$  (see Fig. 1). For such a transformation, the following coordinate equations are valid:

$$\sin \theta' \cos \varphi' = \sin \theta \cos \varphi \cos \theta_{nm} - \cos \theta \sin \theta_{nm},$$

$$\sin \theta' \sin \varphi' = \sin \theta \sin \varphi,$$

$$\cos \theta' = \sin \theta \cos \varphi \sin \theta_{nm} + \cos \theta \cos \theta_{nm}.$$

The prime sign here stands for the spherical angles in the new coordinate system. With the allowance made for an obvious condition that  $\varphi' = \pi/2$ , these equations transform into the equation relating the polar angles  $\theta$  and  $\theta'$ :

$$\cos \theta = \cos \theta' \cos \theta_{nm}.$$

Then we finally obtain

$$q_{nm} \approx 1/2 (m^2/n^2)$$

for spheroidal deformations, and

$$q_{nm} \approx 1/2 \bar{\xi}_A^2 (m^2/n^2)$$

for combined deformations ( $N = 1$ ).

These equations show that degeneration by the azimuth index (in the meaning of natural frequencies) is lifted in the presence of deformations of a spherical particle, and the absence of changes in the Q-factor of the resonance modes with  $m = 0$  is a consequence of the absence of surface deformations at  $\theta_{nm} = \pi/2$  [Eqs. (13) and (15)].

Thus, surface deformation of particles causes a kind of selection of the excited WG modes, that is of primary importance for, for example, support of stimulated scattering processes. It is obvious that in an ideal sphere the competition of resonance modes is always won by the highest-Q electromagnetic oscillations having the lowest order. But the situation drastically changes, if the surface of a liquid particle is deformed. In this case, the modes having the lower Q-factor, but higher stability to deformations of the particle surface come over.

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