Surface and internal scattering in microspheres: limits imposed on the Q-factor and mode splitting

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Abstract

Accurate calculation of internal and surface scattering losses in fused silica microspheres is done. We show that in microspheres internal scattering is partly inhibited as compared to losses in the bulk material. We pay attention on the effect of frozen thermodynamical capillary waves on surface roughness. We calculate also the value of mode splitting due to backscattering and other effects of this backscattering.

Optical microsphere resonators working on whispering gallery modes combine several unique features – very high quality factor, small size and effective volume of field localization with low cost, that make them very attractive for future applications in optoelectronics and measurement science. Such microresonators may be for example used as interferometers and filters with record finesse for QED experiments and for diode laser stabilization. The last application as for now looks the most intriguing as it allows to create centimeter size cheap tunable laser with kilohertz linewidth.

Microspheres are prepared from fused silica or other pure glassy medium by autoforming under the action of surface tension in the flame of a microburner or in the CO₂ beam.

The properties of the microspheres are analyzed theoretically and experimentally rather wide. The theory of whispering gallery modes is well known from electrodynamics and methods of optimal coupling with them were elaborated and experimentally confirmed. In nonlinear properties of fused silica microspheres were investigated theoretically and experimentally. Mechanisms limiting the quality factor of microspheres were outlined in several papers. It was shown that the main factor preventing the obtaining and preservation of high Q-factor is surface atmospheric water adsorption. However, values as high as \( Q \approx 8 \times 10^9 \) – very close to fundamental limit of internal losses were obtained at He-Ne and near IR region. Attempts to obtain higher Q moving to minimum of losses of fused silica 1.55\( \mu \)m, were as for now unsuccessful. Besides chemosorbed water the reason may be surface scattering on inhomogeneities.

Problems of scattering inside the microsphere and on its surface were analyzed formerly incompletely. Scattering leads not only to the limitation of the Q-factor but also lifts degeneracy between degenerate sine and cosine modes. This effect may be observed as mode doublets. Especially badly explored are questions of surface scattering. In different papers one may find expressions leading not only to different numerical estimates but to different functional dependencies from the size of the resonator and wavelength.

1. Scattering on internal thermodynamical inhomogeneities and quality-factor of microspheres

Intrinsic scattering and absorption losses in microresonators were estimated previously from the bulk losses as:

\[
Q = \frac{2\pi n}{\alpha \lambda},
\]

where \( n \) is index of refraction \( \alpha \) is intensity attenuation coefficient and \( \lambda \) is wavelength. However, this approach is not quite accurate for scattering. We remind here the method of derivation of scattering coefficient \( \alpha \) (see for example) to see what modifications should be made in Eq. (1) to take into account specific features of the microspheres.

Let us divide the whole volume of medium in small volume \( dv \), each having due to fluctuations dielectric constant \( \epsilon(\vec{r}) = \delta \epsilon(\vec{r}) + \epsilon^0 \). Internal small inhomogeneities in the field of the mode behave as dipoles reradiating light in all direction according to the Rayleigh formula:

\[
\frac{I_s}{T} = \frac{\pi^2 \sin^2 \theta}{\lambda^4 r^2} \left( \int \int \delta \epsilon(\vec{r}_1) \delta \epsilon(\vec{r}_2) dv_1 dv_2 \right),
\]
where \( \vartheta \) is the angle between the dipole axis (coinciding with polarization of field) and direction of scattering, \( r \) is the distance from scatterer.

The next step is to integrate Eq. (2) over all angles on large sphere \( (r \to \infty) \) to obtain total power of scattering.

\[
P_s = I \frac{8 \pi^2}{3 \lambda^4} \int \int \delta(\vec{r}_1) \delta(\vec{r}_2) dv_1 dv_2,
\]

(3)

However, for the microsphere this is not correct. We should take into account total internal reflection (TIR).

Beams falling on the surface under the angle larger than critical one \( \gamma_0 = \arcsin(1/n) \) will either go back in the mode if this angle lies inside the mode’s caustic or will be suppressed in destructive interference during several reflections. These beams may also go to another mode, which leads to internal mode coupling, but due to the rareness of mode spectrum this effect is negligible (specific case of coupling between oppositely circulating degenerate modes is analyzed below separately). In this way, only beams falling under angles less than critical should be added to losses. We may not take into account here Fresnel transmission coefficients as these beams may leave the resonator during several reflections. Conditions for the cutting of angles for \( TE \) and \( TM \) modes are the following:

\[
\sin^2 \gamma_{TE} = \left( \frac{a - d}{a} \right)^2 \left( 1 - \sin^2 \vartheta \cos^2 \varphi \right) < \frac{1}{n^2}
\]

\[
\sin^2 \gamma_{TM} = \left( \frac{a - d}{a} \right)^2 \sin^2 \vartheta < \frac{1}{n^2},
\]

(4)

here \( d \) is the distance of the dipole from the surface and \( a \) is the radius of the microsphere. If \( d \ll a \), that is always correct for high-Q whispering gallery modes, the first terms in (4) may be omitted and hence the result will not depend from the size of the resonator. We skip here the following derivation of \( \alpha \) by calculating thermodynamical calculations as conditions of angle cutting for large microspheres do not interfere with it (see 12).

\[
\alpha_{is} = \frac{8 \pi^3}{3 \lambda^2 n^8 p^2 \kappa T \beta_T},
\]

(5)

where \( \kappa \) is the Boltzman constant, \( T \) is the effective temperature of glassification \((\sim 1500 K \) for fused silica), \( \beta_T \) is isothermic compressibility, and \( p \) is Pokkels coefficient of optoelectricity at this temperature.

Cutting conditions in this way may be taken into account by introducing suppression coefficients:

\[
Q_{is} = K_{TE,TM} \frac{2 \pi n}{\alpha_{is} \lambda},
\]

(6)

This coefficient \( K_{TE,TM} \) is equal to the relation of complete scattered power to the power scattered on angles satisfying conditions (4).

Numerical calculations for fused silica with \( n = 1.45 \) give

\[
K_{TE} = 2.8 \quad K_{TM} = 9.6
\]

(7)

It follows from these values that TM-modes are less sensitive to intrinsic scattering losses, but these modes have stronger field on the surface, and therefore more sensitive to surface inhomogeneities and absorption on surface contaminations.

2. Scattering on surface roughness

To analyze surface scattering we analogously to the previous section calculate the value of \( \alpha_{ss} \), describing losses of travelling wave per unit length. We start with the same expression in integral form (2), but now shall take into account only surface inhomogeneities. As before we should integrate this expression over angles with account of TIR, but for surface dipoles the part of light scattered above the surface may go free. In this way, suppression coefficient may be taken as \( 2K_{TE,TM}/(K_{TE,TM} + 1) \). In calculations of attenuation coefficient we as above for generality are not taking it into account and insert only in final formula for the quality-factor.

Let the wave with intensity distribution \( I(y, z) \) travels along a guiding surface along the local \( x \)-axis, \( y \)-axis is chosen also along, and \( z \)-axis orthogonally to the surface. Small surface roughness leads to inhomogeneity of dielectric constant:

\[
\delta \epsilon(x, y, z) = (\epsilon_0 - 1) f(x, y) \delta(z),
\]

(8)
here $\delta(z)$ is delta function. If surface inhomogeneities are weakly correlated and their correlation function quickly abates to zero on the scale much smaller than the wavelength, roughness may be described by only two parameters – variance $\sigma = \sqrt{\langle f(x,y)^2 \rangle}$ and correlation length $B$. In this case

$$P_s = dx \int I(y,0) \frac{16\pi^2}{3\lambda^4} (n^2 - 1)\pi B^2 \sigma^2 dx = P\sigma dx,$$  

(9)

Thus, considering that the power of the wave is equal to $P = \int I(y,z) dy dz$, and considering that the wave travels close to the surface, we obtain:

$$\alpha_{ss} = \frac{I(y,0)}{\int I(y,z) dz} \frac{16(n^2 - 1)\pi^3 B^2 \sigma^2}{3\lambda^4}.$$

(10)

Now we turn back to microsphere to calculate the ratio of intensities in above expression. For simplicity we limit ourselves only with TE-mode, which usually have higher quality factor. As the intensity is proportional to the square of electric field, and fields are described by Bessel functions, using the same approximations as in we obtain for TE-mode:

$$\int_0^a \frac{j_\ell^2(knr)r^2 dr}{a^2 j_\ell^2(kna)} \simeq \frac{a}{2j_\ell^2(kna)} \left( \frac{\partial j_\ell(\rho)}{\partial \rho} \right)_{\rho=kna}^2 \simeq \frac{a(n^2 - 1)}{2n^2},$$

(11)

Finally we obtain expression for the quality factor:

$$Q_{ss} = \frac{K_{TE}}{1 + K_{TE}} \frac{3\lambda^3 a}{8\pi n^2 B^2 \sigma^2}.$$

(12)

We may note that this expression is different from expression obtained in but the reason is that the authors underestimated volumetric ratio considering it proportional to $\sqrt{a\lambda}$ and not $a$. In the same paper authors report on measurement of surface roughness of fused silica by means of scanning tunneling microscope. Values $B = 5nm$ and $\sigma = 1.7nm$ were estimated. On Fig.1 all calculated limitations on quality factor are plotted, on this graphics UV and IR absorption in fused silica from literature are also taken into account. Bulk losses in fused silica were taken as:

$$\alpha \simeq (0.7 \mu m^4/\lambda^4 + 1.1 \times 10^{-3} \exp(4.6\mu m/\lambda) + 4 \times 10^{12} \exp(-56\mu m/\lambda)) dB/km$$

(13)
For the estimates of surface scattering resonator radius $a = 300 \mu m$ was taken. One can see that for the measured $Q$ in He-Ne range suppression of scattering is compensated by surface scattering. For longer wavelengths, it seems, additional experiments are required. For very large spheres (several millimeters in diameter), quality factor sufficiently higher then $10^{11}$ may be obtained. It seems that even $Q \approx 10^{12}$ is not impossible as Rayleigh scattering may be lowered by heat treatment on $25\%$. The only unknown factor is surface absorption on hemosorbed layers of $OH^-$ ions and water. It should scale linearly with $a$ as surface scattering (estimate in [3] is also based on the wrong volumetric ratio and in this way incorrect) but its dependence on $\lambda$ is unknown. It is known, however, that in the bulk hydroxil ions lead to vibronic absorption peak at $\lambda = 2.73 \mu m$ and at overtones at $1.37$, $0.95$, $0.725$ and $0.586 \mu m$.

Nevertheless the problem of surface scattering in microspheres is not yet closed neither theoretically nor experimentally. It is reasonable to suggest that surface roughness may be attributed to surface capillary waves frozen during solidification. These waves leads to fluctuations:

$$f(\theta, \phi) = \sum_{L,M} b_L Y_L^M(\theta, \phi),$$

where $Y_L^M$ is spherical angular function, and $L > 1$. If according to thermodynamics each wave has energy $\kappa T$, then

$$< b_L^2 >= \frac{\kappa T}{\tilde{\sigma}(L-1)(L+2)},$$

where $\tilde{\sigma}$ is the coefficient of surface tension $\sim 200$ dyn/cm for fused silica at temperature $T = 1500 K$. Though as estimates show the size of these fluctuations will be several times less than measured in [3] correlation function, calculated for such inhomogeneities has logarithmic shape and in this way may not be characterized by correlation length. Fluctuations on the scale of wavelength are of the same order as in nanoscale. In other words, our approach will not work for this case. Unfortunately the estimate done in [4] for scattering on capillary waves $Q_{cs} \approx a\lambda/((n^2 - 1)b_L^2)$ is also incorrect (it was shown in [5] and experimentally confirmed in [6] that perturbation with $M=0$ (ellipticity if $L=2$), do
not perturb in the first order of approximation quality factor at all). The problem of frozen capillary waves in any case deserves special consideration.

3. Coupled modes in microspheres

Mode coupling in microspheres due to surface and internal inhomogeneities may be described using variational approach. Random deviations of dielectric constant may be written in the form:

$$\delta \epsilon = f(\theta, \phi) F(r),$$

where $F(r)$ – is random radial function, and $f(\theta, \phi)$ – is random angular function. In particular case of small surface roughness when random fluctuations of the surface of the sphere may be described as:

$$r(\theta, \phi) = a + f(\theta, \phi),$$

(17)

and expression (16) may be written as

$$\delta \epsilon = (n^2 - 1) f(\theta, \phi) \delta(r - a).$$

(18)

From the Maxwell equation, wave equation for the fields inside the microsphere with inhomogeneities may be obtained:

$$\Delta E - \left( \frac{\epsilon_0^0(r)}{c^2} + \frac{\delta \epsilon(r)}{c^2} \right) \frac{\partial^2 E}{\partial t^2} = 0$$

(19)

The solutions of unperturbed equation without inhomogeneities (if $\delta \epsilon = 0$) have the form

$$\tilde{E}_j = e^{(\omega_j t)} \bar{\epsilon}(r, \theta, \phi),$$

(20)

where $\bar{\epsilon}(r, \theta, \phi)$ – is vector harmonic, satisfying Helmholtz equation:

$$\Delta \bar{\epsilon}_j + \epsilon_0^0 k_j^2 \bar{\epsilon}_j = 0$$

(21)

and index $j$ corresponds to all possible types of oscillations, and $j = 0$ corresponds to initially excited mode. Using the method of slowly varying amplitudes we find solution as:

$$\tilde{E} = e^{-i \omega_0 t} \sum A_j(t) \tilde{\epsilon}_j$$

(22)

After substituting this sum in equation (19) and omitting small terms we obtain:

$$2i\omega_0 \epsilon_0^0 \sum \frac{dA_j(t)}{dt} \tilde{\epsilon}_j + \omega_0^2 \delta \epsilon \sum A_j(t) \tilde{\epsilon}_j + \epsilon_0^0 \sum (\omega_j^2 - \omega_0^2) A_j(t) \tilde{\epsilon}_j = 0$$

(23)

After multiplication of this equation on $\tilde{\epsilon}_j^*$ and integration over the whole volume, with account of modes’ orthogonality, we obtain usual equations for coupled modes:

$$\frac{dA_k}{dt} + i \Delta \omega_k A_k = i \sum_j A_j \beta_{jk},$$

(24)

where $\Delta \omega_k = \omega_k - \omega_0$ and

$$\beta_{jk} = \frac{\omega_0}{2n^2} \int \bar{\epsilon}_j \delta \epsilon \bar{\epsilon}_k^* dv \int |\tilde{\epsilon}_j|^2 dv$$

(25)

In this expression it is the random function $\delta \epsilon$ which leads to the coupling between $\tilde{\epsilon}_j$ and $\tilde{\epsilon}_k$. We are interested only on the modulus of the coefficient of $\beta_{jk}$, which determines the rate of energy redistribution between different modes. If the value of inhomogeneities and their correlation length are very small if compared with wavelength we may average $\beta_{jk}^2$ and obtain that:
\[
\beta_{jk}^2 = \frac{\omega_1^2}{4n^4} \frac{\langle \delta \epsilon(\bar{r}) \delta \epsilon(0) dv \rangle}{V_{jk}},
\]
(26)

where \( V_{jk} \) – is overlap volume of modes:

\[
V_{jk} = \frac{\int |e_j|^2 dv \int |e_k|^2 dv}{\int |e_j|^4 dv}.
\]
(27)

In the most interesting case of coupling between two modes \( A_+ (t) \) and \( A_- (t) \), travelling inside the microsphere in opposite direction, fields’ distribution for these two modes differs only on phase factor \( \exp(\pm im\phi) \). In this way \( \bar{e}_j = \bar{e}_k \) and \( V_{jk} \) in this case transforms in effective volume of field localization:

\[
V_{\text{eff}} = \frac{(\int |e_j|^2 dv)^2}{\int |e_j|^4 dv},
\]
(28)

4. Effects of coupled modes

To analyze the consequences of internal coupling between modes travelling in opposite directions on the output characteristics of a resonator we may use the same quasigeometrical approach that we used in \( \ref{7} \) to analyze coupler devices for the microspheres. For simplicity we analyze here only the case of ideally mode matched (or monomode) coupler device. The set of equations for the internal and external amplitudes looks as follows:

\[
\frac{dA_+}{dt} + (\delta_0 + \delta_c + i\Delta\omega)A_+ = iA_- \beta + i\frac{T}{\tau_0}B_{\text{in}}
\]
\[
\frac{dA_-}{dt} + (\delta_0 + \delta_c + i\Delta\omega)A_- = iA_+ \beta
\]
\[
B_t = \sqrt{1 - T^2} B_{\text{in}} + iT A_+
\]
\[
B_r = iT A_-
\]
(29)

Fig. 2. Backscattering in a microspere

Where \( A_+ (t) \) and \( A_- (t) \) are as before the amplitudes of oppositely circulating modes of TIR in the resonator (Fig. 2) to model the whispering gallery modes. \( B_{\text{in}} \) is the amplitude of pump and \( B_t \) and \( B_r \) are output amplitudes transmitted and reflected in coupler. \( T \) is the amplitude transmittance coefficient, describing coupler, \( \delta_0 = 2\pi n/\alpha \lambda \) is
the decrement of internal losses, $\delta_c = T^2/2\tau_0$ is the decrement of coupler device, $\tau_0$ is the circulation time $\tau_0 \simeq 2\pi na/c$, and $\Delta\omega$ is frequency detuning from unperturbed resonance frequency $\omega_0$ (for details see 7). The stationary solution of Eq. (29) is the following

$$A_+ = i \frac{2\delta_c \beta}{\overline{T}((\delta_0 + \delta_c)^2 + \beta^2 - \Delta\omega^2 + i2\Delta\omega(\delta_0 + \delta_c))} B_{in}$$

$$A_- = -\frac{1}{\overline{T}} \frac{2\delta_c(\delta_0 + \delta_c + i\Delta\omega)}{(\delta_0 + \delta_c)^2 + \beta^2 - \Delta\omega^2 + i2\Delta\omega(\delta_0 + \delta_c)} B_{in}$$

$$B_t = \frac{\delta_0^2 - \delta_c^2 + \beta^2 - \Delta\omega^2 + i2\delta_0 \Delta\omega}{(\delta_0 + \delta_c)^2 + \beta^2 - \Delta\omega^2 + i2\Delta\omega(\delta_0 + \delta_c)} B_{in}$$

$$B_r = -\frac{i2\delta_c \beta}{(\delta_0 + \delta_c)^2 + \beta^2 - \Delta\omega^2 + i2\Delta\omega(\delta_0 + \delta_c)} B_{in}$$

(30)

If internal mode coupling constant is weaker than attenuation $\beta < \delta_0 + \delta_c$ than Eq. (30) has only one resonance at $\Delta\omega = 0$ and backscattering is small. The situation is not very different in this case from the case of one mode analyzed in 7. In temporal language it means that internal coupling simply has no time to build backscattered wave during the ringdown time. Interesting is, however, that the regime of critical coupling (when $B_t = 0$) is shifted and obtained not for $\delta_c = \delta_0$ but for $\delta_c^2 = \delta_0^2 + \beta^2$ and in this case not all input power is lost in the resonator but some part of it reflects back in the coupler.

$$B_r = i \frac{\beta}{\delta_0 + \delta_c}$$

(31)

This means that ringdown time is equal to the time needed to repump circulating mode in oppositely circulating one.

\[ |B_r|^2/|B_{in}|^2 = \frac{\delta_c^2}{\delta_0^2} \]

Fig. 3. The dependence of power, reflected in coupler due to backscattering in microsphere, from loading

$$|A_+|^2 = |A_+|^2 = \frac{1}{\overline{T}^2((\delta_0 + \delta_c)^2 + \beta^2 - \Delta\omega^2 + i2\Delta\omega(\delta_0 + \delta_c))} B_{in}^2$$
\[ |B_t|^2 = \frac{\delta_0^2}{(\delta_c + \delta_0)^2} B_{in} \quad \quad |B_r|^2 = \frac{\delta_c^2}{(\delta_c + \delta_0)^2} B_{in} \]  

(32)

The case of \( \beta \geq \delta_0 + \delta_c \) is much more interesting and even leads to somewhat unexpected results. In this case there are two resonances at frequencies \( \Delta \omega = \pm \sqrt{(\beta^2 - (\delta_0 + \delta_c)^2)} \) i.e. internal coupling lifts degeneracy between sine and cosine standing modes in the microsphere and there is enough time to form them. All intensities in resonances in this case do not depend on \( \beta \):

What is interesting and infeasible that if \( \delta_0 \ll \delta_c \) (overcoupling) but still \( \delta_0 + \delta_c < \beta \) then most part of input power is backscattered and transmitted power tends to zero. This property may become extremely valuable for future applications of microspheres in laser stabilization. To verify this result additional experiments are required, but we saw many times that backscattering is practically absent when there is no splitting and practically does not depend on loading when doublets are clearly seen (see Fig.3).

5. Calculation of mode splitting on internal and surface inhomogeneities

Mode coupling leads to splitting of initially degenerate modes, if \( \beta \) constant is much larger than mode decrement of internal and coupling attenuation \( \delta_0 + \delta_c \) then:

\[ \frac{\Delta \omega}{\omega} = \frac{2\beta}{\omega_0}, \]

(33)

If thermodynamical inhomogeneities are calculated in the same way as before:

\[ \left( \frac{\Delta \omega}{\omega} \right)_{is} = \sqrt{n^4 p^2 \kappa T \beta \tau} = \sqrt{\frac{3\lambda^4 \alpha_{is}}{8\pi^3 n^4 V_{eff}}}, \]

in agreement with qualitative estimates in. Effective volume of the most interesting \( TE_{\ell\ell_1} \)-mode may be calculated according to the formula:

\[ V_{eff} = 2.3n^{-7/6} a^{11/6} \lambda^{-7/6}, \]

(35)

In this way, for \( TE_{\ell\ell_1} \)-mode in fused silica microsphere:

\[ \left( \frac{\Delta \omega}{\omega} \right)_{is} \approx \frac{5 \times 10^{-7} \mu m^{3/2}}{\lambda^{11/12}a^{7/12}}, \]

(36)

If \( \ell \neq m \) the following asymptotic approximation is valid:

\[ V_{eff,\ell m} = V_{eff,\ell \ell}(1 + 0.5\sqrt{T - m - 0.5}), \]

(37)

Now let us analyze the case of mode splitting due to the surface inhomogeneities. From (28) and (3) after averaging

\[ \beta_{ss}^2 = \frac{\omega_0^2 \pi B^2 \sigma_0^2 |\tilde{c}|^4}{4n^4 V_{eff} \int |\tilde{c}(r)|^4 dr} \]

(38)

or for \( TE_{\ell\ell_1} \)-mode in fused silica microsphere:

\[ \left( \frac{\Delta \omega}{\omega} \right)_{ss} \approx \frac{1.1\sigma B}{\lambda^{1/4} a^{7/4}}, \]

(39)

It is easy to show that for measured size of surface inhomogeneities this expression gives substantially lower level of coupling between modes than internal inhomogeneities. On (Fig. 3) results of calculation for fused silica microsphere for \( \lambda = 0.63 \mu m \) according to (36,39) are shown.
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