Ultimate $Q$ of optical microsphere resonators

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Received November 15, 1995

We demonstrate the quality factor $Q = (0.8 \pm 0.1) \times 10^{10}$ of whispering-gallery modes in fused-silica microspheres at 633 nm, close to the ultimate level determined by fundamental material attenuation as measured in optical fibers. The lifetime of ultimate $Q$ is limited by adsorption of atmospheric water. Monitoring of adsorption kinetics with submonolayer sensitivity by $Q$ factors and frequencies of whispering-gallery modes is demonstrated. The possibility of supermaterial $Q$‘s owing to intrinsic suppression of scattering losses in microspheres is discussed. © 1996 Optical Society of America

Microspheres of fused silica with high-$Q$ whispering-gallery (WG) modes (hereafter called microspheres)\textsuperscript{1–3} are a novel type of optical resonator that are attractive for different applications. Most applications, especially projected cavity QED experiments,\textsuperscript{4,5} require the realization of the highest possible $Q$. Since its first demonstration in 1987, microsphere $Q$ has been improved from nearly $1 \times 10^6$ to $(2–3) \times 10^9$ in the red and near-IR areas of spectrum.\textsuperscript{3,5} This $Q$ was, however, at least three times smaller than the limit determined by intrinsic material losses. It was also reported that in regular laboratory conditions the $Q$ factor deteriorates within a 1-h time scale, presumably because of deposition of microdust or water vapors upon the microsphere surface.\textsuperscript{5} Thus far it has remained unclear whether a material-limited $Q$ can be realized in experiment.

The quality factor of WG modes is determined by several factors:

$$Q^{-1} = Q_{\text{rad}}^{-1} + Q_{\text{s.s.}}^{-1} + Q_{\text{cont}}^{-1} + Q_{\text{mat}}^{-1},$$

where $Q_{\text{rad}}^{-1}$ denotes intrinsic radiative (curvature) losses; $Q_{\text{s.s.}}^{-1}$, scattering losses on residual surface inhomogeneities; $Q_{\text{cont}}^{-1}$, losses introduced by surface contaminants; and $Q_{\text{mat}}^{-1}$, material losses. As frequently described, $Q_{\text{rad}}^{-1}$ vanishes exponentially with increasing size, so with $D/\lambda \geq 15$, $Q_{\text{rad}} > 10^{11}$ ($D$ is the microsphere diameter; $\lambda$ is the wavelength). $Q_{\text{s.s.}}^{-1}$ can prevail in intermediate-sized spheres. Calculations based on the model of Rayleigh scattering by molecular-sized surface clusters under grazing incidence and total internal reflection yield the following estimate for $Q_{\text{s.s.}}^{-1}$:

$$Q_{\text{s.s.}} = \frac{\lambda^2 D}{2\pi^2 \sigma^2 B},$$

where $\sigma$ and $B$ are the rms size and the correlation length of surface inhomogeneities, respectively. With the numerical values $\sigma = 0.3 \text{ nm}$ and $B = 3 \text{ nm}$ reported for glass surfaces\textsuperscript{8} we obtain that $Q_{\text{s.s.}}^{-1} \ll 1 \times 10^{-10}$ may be expected only in large spheres, $D \gg 100 \mu m$. In the absence of contaminants the $Q$ of large spheres may reach the limit defined by material losses. Optical attenuation in fused silica, investigated intensively in the context of fiber transmission optimization, at 633 nm is $\approx7 \text{ dB/km}$ (5 dB is bulk Rayleigh scattering and 2 dB is absorption).\textsuperscript{7} Therefore the principal limit for microsphere $Q$ at 633 nm is

$$Q_{\text{mat}} = \frac{2\pi n}{\alpha \lambda} = 0.9 \times 10^{10}.$$
each time between the 50th and the 60th second after fabrication.

Figure 2(a) shows a typical time dependence of $Q$, with the $t = 0$ point coinciding with the fabrication of microsphere (its removal from the flame after formation of sphere by surface tension forces). The $Q$-versus-$t$ plot indicates quick decay of the record $Q$ within the first $5$ min toward $\sim 20\%$ of the record $Q$ and slower saturation toward $Q \approx 1 \times 10^9$ with a much longer lifetime on a many-hour scale. Bakeout of the resonator at $400\,{}^\circ C$ for $30$ s resulted in partial restoration of $Q$ [Fig. 2(b)], in favor of the water adsorption hypothesis.

To obtain additional information on the kinetics of surface contamination, we also measured the time variation of WG mode frequencies [Fig. 2(c)]. We provided a stable reference for measurement of the shift of WG mode frequencies by beating the probe laser light with an additional frequency-stabilized He–Ne laser.

Resolution of the measurement was limited by the temperature drift of the microsphere in passive heat isolation (total drift of the frequency of a selected mode in a saturated microsphere $5$ h after fabrication smaller than $1$ MHz for $20$ min).

According to a current model of two-stage chemosorption, after fast adsorption of oxygen upon a fresh $\text{SiO}_2$ surface the following hemosorption of atmospheric water leads to formation of a layer of OH groups chemically bound to the surface. The reported duration of this process, of the order of $100$ s, agrees both with the parameter of logarithmic fit of the $\Delta \nu$-versus-$t$ plot [solid curve in Fig. 2(c)] and with the characteristic time of the initial fast drop of $Q$ in our experiment. The resulting hydrated surface is a substrate for further adsorption of molecular water. After this stage, presumably completed in $20–30$ min, only slow deposition of microdust particles is responsible for long-term degradation of $Q$. (In a hermetic box, we have observed $Q \approx 1 \times 10^9$ of a $350-\mu m$ resonator preserved during $t \geq 6$ months.)

The results of measurement of WG mode frequency variation can be interpreted in terms of the thickness $\delta$ of an adsorbed layer:

$$\delta = \frac{\Delta \nu n_{\text{ads}}^2 - 1}{\nu n_{\text{sil}}^2 - 1} \frac{D}{2} \approx \left(\frac{\Delta \nu [\text{MHz}]}{800}\right) [\text{nm}],$$

where $\Delta \nu$ is the current-frequency shift, $\nu \approx 475$ THz is the optical frequency, $n_{\text{sil}}$ and $n_{\text{ads}}$ are refractive indices of silica and of the adsorbed layer, respectively, a numerical estimate is given for $n_{\text{sil}} = 1.46$, $n_{\text{ads}} = 1.33$, and $D = 750$ $\mu m$. According to results shown in Fig. 1(c), the total change in thickness of the adsorbed layer between $1$ and $30$ min is $\sim 0.2$ nm and corresponds to $1.5$ monolayers.

Extrapolation of the results in Figs. 1 and 2 (to estimate the quality factor of a fresh microsphere) cannot be based on the logarithmic fit of the $\Delta \nu$-versus-$t$ plot. The kinetics of the initial stages of adsorption near $t = 0$ can be different and requires specially arranged measurements close to the formation time, with attention to the possibility of laser-induced effects. (In our experiments the time interval between formation and first measurement points was limited by the transfer of the ready microsphere from the torch to the optical table and by alignment of the setup). However, the absence of an apparent shelf at the level $Q_{\text{mat}}$ in a $Q$-versus-$t$ curve may indicate that $Q > Q_{\text{mat}} = 0.9 \times 10^{10}$ can be expected at $t = 0$. We cannot rule out this possibility because scattering losses in microspheres can be smaller than in bulk material or fibers, in which the lowest attenuation in fused silica has been measured.

Indeed, the Rayleigh losses in fused silica are attributed to scattering of an electromagnetic wave on thermal and frozen fluctuations of density. For a free traveling wave this mostly paraxial scattering produces irreversible losses of energy by coupling the initial wave to the radiative modes of space. In a microsphere, because of its narrow angular distribution, this scattering must predominantly couple the initial mode to other close-frequency WG modes with similar configuration. However, this process is suppressed because of the rare spectrum and high $Q$ of the modes. The only modes that efficiently couple to each

Fig. 1. Mode energy damping curve for a WG mode in a $750-\mu m$ sphere. Estimated damping time $\tau = 2.7$ $\mu s; \lambda = 633$ nm.

Fig. 2. (a), (b) Effect of adsorption of atmospheric water on the damping time and (c) frequency of a WG mode in a $750-\mu m$ microsphere. (b) Illustrates the effect of $30$-s bakeout at $400\,{}^\circ C$.
other are originally degenerate opposite-rotating high-
Q WG modes. This effect of backscattering results not
in losses but in the splitting of resonances reported
earlier.\textsuperscript{11,12}

Independently, larger $Q$ can be expected closer to the
minimum of attenuation in fused silica, promising at
least $1.5 \times 10^{11}$ at $\lambda = 1.55$ $\mu$m. And, apparently, to
preserve the record $Q$ for applications, microspheres
have to be prepared and contained in evacuated or dry-
gas-filled chambers. Another possibility for preserva-
tion of the record $Q$ is chemical treatment to prevent
surface hydration in the atmosphere.

This research has been performed as part of collabora-
tive project with the University of California and
the Los Alamos National Laboratory and supported in
part by the Quantum Optics Lab, California Institute
of Technology. The authors are grateful to V. B.
Braginsky, H. J. Kimble, H. Mabuchi, and S. Habib for
helpful discussions.

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