High-resolution spectroscopy of whispering gallery modes in large dielectric spheres

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The mode spectrum of a 3.8-cm-diameter fused-silica sphere has been studied in the vicinity of 1.06 μm. A single-frequency Nd:YAG laser was used to excite whispering gallery modes by means of evanescent wave coupling. The spectrum is in excellent agreement with predictions from Mie theory.

The interaction of optical waves with spherical objects has been a subject of interest since the early treatments by Lorenz, Mie, and Debye,[1] which provide the framework for the description of various natural light-scattering phenomena. More recently, investigations have been extended to nonlinear-optical interactions in small spheres,[2] which are favored by the strong spatial confinement of resonance modes. The observation that a spherical object can sustain high-Q resonator modes dates back to Richtmyer.[3] Spherical resonators for optical waves have been used as laser resonators.[4] Mie theory[1] predicts that the modes' radial spatial extension from the surface toward the center of the sphere lies in the range from a few wavelengths to $(1 - n_a/n_s)R$, where R is the sphere's radius and n_s and n_a are the refractive indices of the sphere and the medium surrounding it. The former modes, called whispering gallery modes, have been observed in stimulated-emission,[5] optical-levitation,[6] and fluorescence experiments.[7] Direct excitation by means of a coupling prism has recently been demonstrated by Braginsky *et al.*[8] in fused-silica minispheres using a He–Ne laser.

Here we report detailed results on the resonance frequencies of whispering gallery modes of large dielectric spheres at the Nd:YAG wavelength $\lambda_0 = 1.064$ μm.

Two types of mode, TM and TE, can exist in a spherical resonator; in the limit of resonators large compared with the wavelength of the modes, their electric fields are approximately radially and azimuthally polarized, respectively.[9] The electric fields of the eigenmodes are given by

$$E_r(r,\theta,\phi) \sim \frac{n(n+1)}{kr} j_n(kr) Y_n^q(\theta,\phi) \qquad \text{TM modes,}$$

$$E_\theta(r,\theta,\phi) \sim -\frac{q}{\sin\theta} j_n(kr) Y_n^q(\theta,\phi) \qquad \text{TE modes,} \tag{1}$$

where $k = 2\pi n_s \nu/c$ and j_n, Y_n^q ($q = -n, \ldots, n$) are the spherical Bessel and harmonic functions. Resonance occurs when the scattering coefficient associated with a particular mode n is unity.[10] This is satisfied for an infinite number of reduced eigenfrequencies $n_s x_n^{(l)} = kR$, where the order number l gives the number of radial maxima of the field inside the sphere. High-Q, quasi-bound (i.e., weakly radiating) modes require that $n > n_a kR/n_s$. The small-l modes are the whispering gallery modes, and for large spheres ($R \gg \lambda_0$) their resonance frequencies can be obtained using expansions of the Bessel functions for large order n,[11]

$$\begin{aligned} n_s x_n^{(l)} = n &+ \frac{1}{2} - \zeta_l \left(\frac{n+1/2}{2}\right)^{1/3} - \frac{mp}{\sqrt{m^2-1}} \\ &+ \frac{3\zeta_l^2}{2^{2/3}10(n+1/2)^{1/3}} \\ &- \frac{m^3 p(2p^2/3 - 1)\zeta_l}{2^{1/3}(m^2-1)^{3/2}(n+1/2)^{2/3}}, \end{aligned} \tag{2}$$

where $[x_n^{(l)}, p] = [a_n^{(l)}, 1/m^2], [b_n^{(l)}, 1]$ for TM and TE resonances, respectively, ζ_l denotes the lth zero of the Airy function $\mathrm{Ai}(\zeta)$,[12] and $m = n_s/n_a$.[13]

Here only the first five terms are relevant, and they can be given a simple interpretation. The m-independent terms result from the approximate vanishing of the mode's electric field at the surface of the sphere, $j_n[n_s x_n^{(l)}] \simeq 0$, when in resonance. The mode number n is nearly equal to the number of wavelengths λ_0/n_s that fit around the sphere's circumference, and the free spectral range follows as

$$\nu_f = \frac{c}{2\pi R}[x_{n+1}^{(l)} - x_n^{(l)}] \simeq \frac{c}{2\pi n_s R}. \tag{3}$$

This is the free spectral range expected for a wave that propagates along the inner surface of the sphere that is continuously internally reflected. The total-internal-reflection (TIR) process also gives rise to the frequency shift between the nearly identical TM and TE spectra,

$$\hat{\nu}_p = \frac{a_n^{(l)} - b_n^{(l)}}{\nu_f} \simeq \frac{\sqrt{m^2-1}}{m}, \tag{4}$$

because of the relative phase shifts experienced by the fields on TIR.[14]

While the free spectral range and the frequency shift are relatively insensitive to the precise value of m, the relative positions of the modes of each polar-

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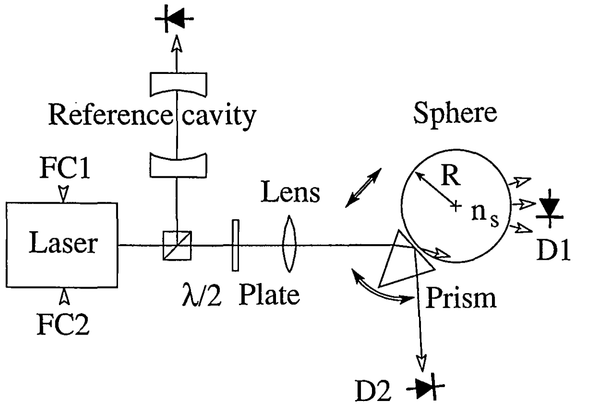

Fig. 1. Setup for the study of modes of spherical resonators. FC1, FC2, thermal and piezoelectric frequency control inputs. D1, D2, alternate positions for the photodetectors that register scattering and outcoupling of the mode field, respectively.

ization are strongly m dependent owing to the large value of n (approximately 10^5 in the present experiment) and can thus be used for an accurate determination of the value of the refractive index.

The apparatus used to investigate the whispering gallery mode spectrum is shown in Fig. 1. An uncoated fused-silica sphere of radius $R = 1.9004$ cm, with deviations from perfect sphericity of less than $\lambda_0/40$, was used. A single-mode, frequency-tunable nonplanar Nd:YAG ring laser[15] with excellent frequency stability properties (10-kHz linewidth) was employed to scan over several free spectral ranges of the sphere. After focusing by a 10-cm focal-length lens, the beam is coupled equatorially into sphere modes by frustrated TIR using a prism. Its refractive index $n_p = 1.69 > n_s$ allows the laser beam to be launched nearly tangentially to the inner surface of the sphere. The sphere is mounted on a translation stage that permits accurate control of the gap distance between coupling prism and sphere by means of a piezoelectric actuator. The TIR modes radiate owing to scattering as well as outcoupling by the same prism used for excitation. When the laser frequency is in resonance with an eigenmode, a ring of light around the equator is observable, caused by surface scattering. The leakage of resonator modes through the coupling prism gives rise to interference with the laser beam internally reflected in the prism. Spectra can be taken by measuring either effect.

Figures 2(a) and 2(b) show the spectra for horizontal (equatorial) and vertical polarization of the laser beam, together with the frequency markers of a Fabry–Perot interferometer, taken by thermally scanning the laser frequency. For comparison, a fit based on the mode spectrum [Eq. (2)] is shown in Fig. 2(c). The agreement with the experimental spectrum is good. An interesting feature is the presence of the lowest-order ($l = 1$) mode, which is usually not observed in scattering or optical-levitation spectra of small particles owing to its small contribution to the scattering cross section.[10]

The mode number is obtained by fitting to 18 of the 22 lowest-order modes, $n = 162554 \pm 1$, with a rms deviation of 1 MHz between data and fit. With a laser frequency $1/\lambda_0 = 9394.0 \pm 0.3$ cm^{-1} and the radius of the sphere, Eq. (2) yields $n_s = 1.45008 \pm 0.00005$.

The measured $\nu_f = 1726 \pm 10$ MHz and polarization shift $\hat{\nu}_p = 0.729$ also compare well with the values of 1731 MHz and 0.724 predicted from Eq. (2) and the fitted index. Temperature drift accounts for the slight discrepancy (tuning coefficient $d\nu/dT \simeq -2$ GHz/K).

To determine accurately the linewidths of the resonances we have studied the derivative line shapes by piezoelectrically frequency modulating the laser and lock-in detecting the equatorially scattered light. The smallest linewidth (FWHM) measured was 3.0 MHz, which implies a resonator finesse $\mathcal{F} \simeq 570$. Modification of the coupling strength by changing the gap width, thereby changing the line intensities over 2 orders of magnitude, did not result in any appreciable change in linewidth. Thus the finesse is due to bulk and surface losses rather than output coupling losses.

With careful material choice and surface polish, fused-silica TIR resonators may reach submegahertz linewidths. This suggests their use as monolithic resonators for frequency stabilization, provided that their operating temperature is tightly controlled. To demonstrate this, the laser was frequency locked to one particular whispering gallery mode, with the derivative signal used as an error signal fed back to the piezoelectric frequency control.

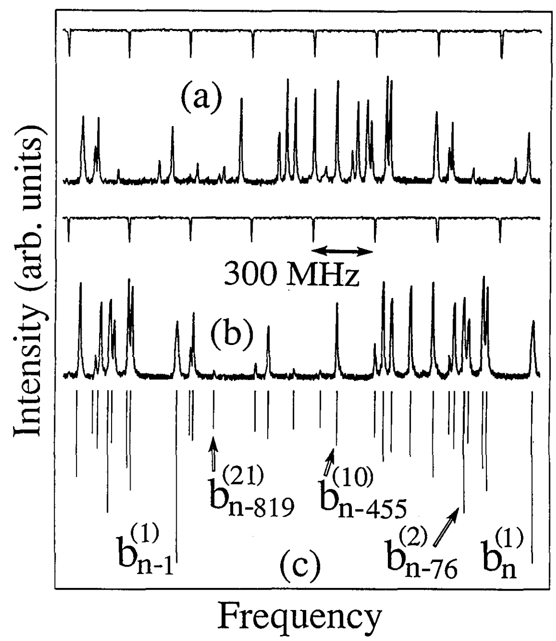

Fig. 2. Experimental and theoretical whispering gallery mode spectra. (a), (b) The intensity of the light scattered from the equator, obtained by lock-in detection, versus the laser frequency together with Fabry–Perot resonances. The scan time was 20 s each. (a) Parallel (in-plane) laser polarization excites TM resonances. (b) TE modes excited by perpendicular laser polarization. (c) The line positions according to Mie theory [Eq. (2)] for l = 1–22, using the best-fit mode number $n = 162554$. For clarity the heights of the bars were scaled by $1/\sqrt{l}$. The center frequency is approximately 9394 cm^{-1}.

The relative intensities of the whispering gallery resonances depend on their excitation probability, i.e., their spatial overlap with the Gaussian laser beam, and their losses. For the scattering spectrum this is a function of the mode's intensity distribution as well as the sphere's scattering loss distribution. The electric-field amplitudes [relations (1)] for the TM and TE whispering gallery modes can be approximated in the large-n case by[11]

$$E(r) \sim Ai\left[\zeta_l + \left(\frac{2k^2}{R}\right)^{1/3}(R - r)\right]. \tag{5}$$

This Airy mode structure has been observed in bent metallic waveguides.[16] Relation (5) implies that the radial depth of the whispering gallery field distribution is $\Delta r \simeq 5|\zeta_l|$ μm in the present case. Since the modes spread out further radially with increasing order number, a decreasing excitation probability is expected for an input beam launched at grazing incidence into the sphere, in agreement with Figs. 2(a) and 2(b), where only the lowest orders appear. With increasing beam launch angle, we observed a growing number of modes as well as strong changes in their intensities.

Similarly we can deduce that the angular distribution must be described by a spherical harmonic peaked near $\theta = \pi/2$, i.e., with q on the order of n, to provide good spatial overlap with the laser beam. Also, according to relations (1), E_θ and E_r are then of the same magnitude, as observed. The presence of the narrow equatorial radiation ring confirms this reasoning (see Refs. 4 and 8).

In conclusion, we have reported the observation and identification of whispering gallery modes of spherical optical resonators using a single-frequency laser. The distinctiveness of the resonances allows laser frequency locking, which is necessary for the implementation of spherical gyroscopes[4] or injection-seeded spherical lasers as well as for the study of nonlinear-optical interactions in resonators such as quantum nondemolition[8] and squeezing effects.[17] Optical spheres are one realization of TIR resonators with evanescent wave coupling. The ability of such resonators to operate over a wide wavelength range with variable input–output coupling promises important applications.

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