Eur. Phys. J. D 1, 235–238 (1998)

THE EUROPEAN
PHYSICAL JOURNAL D

Rapid Note

Evidence for intrinsic Kerr bistability of high-Q microsphere resonators in superfluid helium

F. Treussart[1], V.S. Ilchenko[2], J.-F. Roch[1], J. Hare[1], V. Lefèvre-Seguin[1,a] J.-M. Raimond[1], and S. Haroche[1]

[1] Laboratoire Kastler Brossel, Département de Physique de l'Ecole Normale Supérieure, 24 Rue Lhomond, 75231 Paris Cedex 05, France
[2] Department of Physics, Moscow State University, Moscow 119899, Russia

Received: 16 December 1997 / Accepted: 14 January 1998

Abstract. Quality factors up to 10^9 have been obtained in the whispering gallery modes (WGMs) of fused silica microspheres immersed in a superfluid helium bath. We have observed a dispersive bistable behaviour of the WGM resonances with a threshold power of 10 μW, due to the intrinsic Kerr nonlinearity of silica. These results open the way to the realization of a thresholdless microlaser and other cavity QED projects with microspheres.

PACS. 42.50.-p Quantum optics – 42.55.Sa Microcavity and microdisk lasers – 42.65.-k Nonlinear optics

The whispering gallery modes (WGMs) of silica microspheres are attractive optical resonators for different applications [1–3], going from proposed fundamental Cavity Quantum Electrodynamics (CQED) experiments [4] to more practical systems such as the realization of new kind of microlasers [5]. Light in such WGM's circles around the sphere by repeated total internal reflection at nearly grazing incidence and is strongly confined near the surface in mode volumes of the order of hundreds of μm^3.

In a low-loss material like silica, the quality factor of these WGMs can reach very high values and Q-factors greater than 10^9 have been achieved in various-sized spheres with diameters ranging from 50 to 1000 μm [1,2,6]. Even higher values close to 10^{10} have been obtained on large spheres (diameter 600-1000 μm) during a short period of time following their fabrication [7]. With Q-factors of the order of 10^9, the photon storage time ($\approx 1\mu$s) is much longer than the Rabi period of a one atomic unit dipole coupled to the vacuum field of these modes [8]. This is a favourable condition to observe strong CQED effects. However, thermal effects in the microsphere can impose severe limitations. On one hand, the WGM's suffer from adverse thermal bistability effects due to heating by the probe laser [2,9]. On the other hand, ions or molecules embedded in or coupled to the sphere have their transitions homogeneously broadened by thermal excitations. To reduce these effects, we have developed a cryogenic set-up designed to immerse a microsphere in a superfluid helium bath at 2 K.

We have been able to reproduce at these low temperatures the high-Q factors observed at room temperature. In superfluid helium, the sensitivity of WGM resonances to temperature is greatly reduced and the thermal bistability, easily achieved at 300 K, is no longer observed. As a result, the intrinsic Kerr non linearity of silica becomes dominant and we give evidence below for the observation of this phenomenon.

Our cryogenic set-up is sketched in Figure 1. The WGM's are excited by evanescent-wave coupling using a high index prism (BK7 equilateral prism with 3×3 mm side-faces). The whole mount is placed at the bottom of the inner chamber of a cryostat which is filled with superfluid helium through a sintered copper filter and a capillary tube connected to an external liquid He reservoir. Mechanical feedthroughs are used to compensate for the residual differential thermal contractions and realign the set-up at 2 K. The sphere-prism distance is controlled with nanometric precision. The excitation source is either a 810 nm single-mode diode laser stabilized with an external grating or a free-running DBR laser at 1083 nm [10]. The probe beam is carried by a single-mode optical fiber and focused into the prism by lens L_1 (Thorlabs C220-MP). This objective can be moved in two orthogonal directions by elastic deformations of its mount, so as to align the evanescent field region of the prism in front of the sphere. Resonances are detected on the light coming out of the prism, collected by lens L_2 and coupled into a 100 μm-core multimode fibre, which does not need to be realigned at 2 K.

[a] e-mail: valerie.lefevre@physique.ens.fr

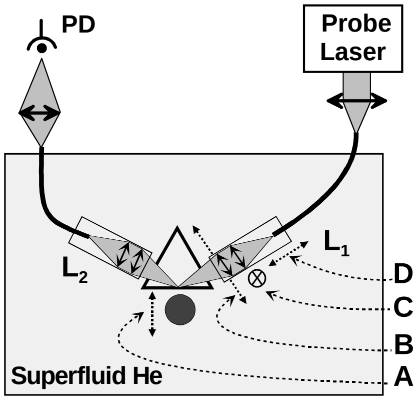

Fig. 1. Cryogenic experimental set-up used to study microspheres immersed in superfluid helium. The WGM resonance is observed on the output beam intensity measured with photodiode PD. The different mechanical motions used to align the sphere and the evanescent field spot at the surface of the coupling prism are shown schematically. Motion A controls the sphere-prism gap (40 μm excursion). Motions B and C are used to change the vertical and horizontal position of lens L_1 (with respect to the mount base). Motion D controls the focusing used to match the WGM transverse profile. These three motions have an excursion range of about 2 mm.

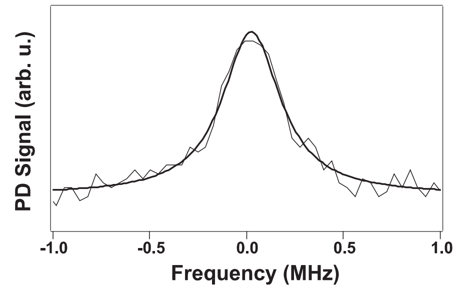

Fig. 2. Observation of a very narrow WGM mode at 810 nm, with a 120 μm silica sphere immersed in superfluid He. The FWMH derived from a lorentzian fit is 390 kHz, yielding $Q = 1.0 \times 10^9$. The frequency calibration of the scan is made by adding a 75 MHz sideband on the probe laser with an AOM.

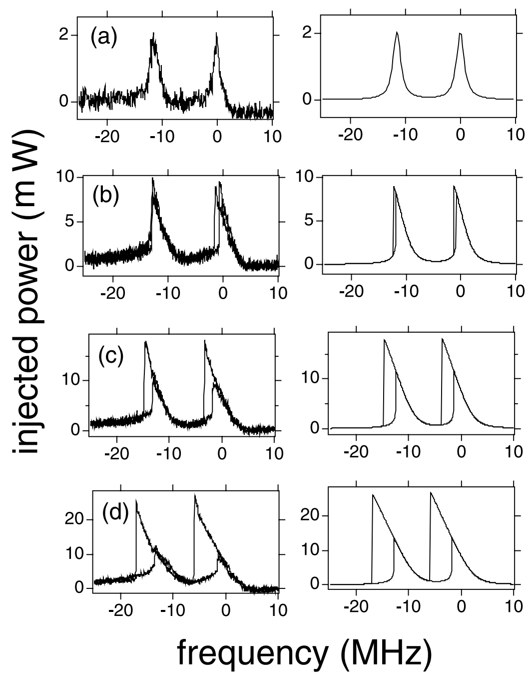

Fig. 3. Observation of a bistable behaviour at 1083 nm on a 50 μm microsphere, for different maximum injected powers P^{inj} in the microsphere ((a) $P^{\text{inj}} = 2$ μW, (b) $P^{\text{inj}} = 9$ μW, (c) $P^{\text{inj}} = 18$ μW, (d) $P^{\text{inj}} = 27$ μW). The quality factor $Q \approx 2 \times 10^8$ is derived from curve (a). The experimental data (on the left side) can be compared with the theoretical curves (on the right side), which are all plotted with the same fit parameter. It gives a threshold power $P^{\text{inj}}_{\text{th}} \approx 10$ μW, consistent with the data shown in (b).

The spheres are made by fusing the end of a high-purity silica wire in a microtorch flame. Great care is taken to protect the microsphere's surface. In particular, we only use clean He gas to thermalize the set-up until superfluid He is admitted in. Screens are also added to shield the sphere from metallic dust particles generated close to the sphere by the motion of the alignment screws.

As a result, we are able to preserve high Q-factors down to 2 K and $Q \geq 10^8$ are achieved routinely on microspheres with 50−100 μm diameter. The WGMs resonances are detected using the transmission of the intracavity field into the prism throughout evanescent wave coupling. When the probe laser frequency is scanned over the WGM resonance, the interference of this output field with the internally reflected field in the prism gives rise to a dip on the output beam intensity. The alignment of the set-up is quite satisfactory since it allows us to reach a maximum coupling efficiency (measured from the dip depth) of the order of 30%. At larger gaps ("undercoupled" regime), the intrinsic Q-factor can be derived from the minimum resonance width. Figure 2 shows a WGM signal observed on a pure silica sphere (120 μm diameter) when the probe laser is scanned over the resonance. A lorentzian fit gives a width of 390 kHz (FWMH), corresponding to $Q = 1.0 \times 10^9$.

As mentioned earlier, we chose to immerse the microsphere in superfluid He to use its enormous thermal conductivity [11] and prevent the thermal bistability effects which are easily obtained at 300 K when scanning back and forth on the WGM resonance [2,9]. With superfluid around the sphere, we nevertheless observed a bistable behaviour on high-Q WGM modes in a 50 μm diameter sphere. It is shown in Figure 3 for a WGM mode split into a doublet by internal backscattering [6], with a threshold injected power $P^{\text{th}}_{\text{inj}} \approx 10$ μW. The observed bistability appears as a negative non-linear frequency shift which can be associated with a positive change of the refractive index of the sphere.

This immediately rules out a thermal effect due to a slight heating of the sphere which has the wrong sign since we have measured a positive thermal frequency shift ($\mathrm{d}\nu/\mathrm{d}T \approx +0.1$ MHz/mK) at 6 K [12].

The data presented here have been taken for various incoming powers with a constant gap, which corresponds to a coupling efficiency of 5%. A numerical fit of the experimental data, based on a frequency shift of the resonance proportional to the power injected in the sphere, is also shown in Figure 3. It can be performed on each component of the doublet independently as long as they are well separated. The Q-factor derived from the lowest power signal (curve (a)), is 2×10^8. The threshold power is given by $P_{\mathrm{th}}^{\mathrm{inj}} = (8\pi/3\sqrt{3})(N^2 V_{\mathrm{M}}/\lambda)(1/n_2 Q^2)$. Taking a typical order of magnitude for the mode volume V_{M} of about 500 μm^3, we derive from our data $n_2 \approx +1.2\times10^{-16}$ cm^2/W, which agrees with the Kerr non-linear refractive index of silica $n_2 = +2.5\times10^{-16}$ cm^2/W [13], considering the experimental uncertainties.

We have made several tests to eliminate possible mimics of the Kerr effect. For instance, a 10^{-6} increase of the refractive index of the superfluid helium located in the WGM evanescent wave would produce the proper negative shift of the WGM resonance (*i.e.* of the order of its width). By changing the temperature of the whole system (He bath and sphere), we have observed negative shifts of the WGMs frequencies of -0.7 MHz/mK well explained by the temperature change of the refractive index of superfluid He [14]. However, the power dissipated in the sphere is not enough to change the temperature of the helium bath around the sphere by 2 mK, which is the temperature change required to shift the resonance by its width. We have also checked that the He density increase due to the electrostriction effect produced by the WGM field cannot explain our data. Moreover, we have studied carefully the bistability effect at several temperatures between 1.8 K and 2.2 K. No change of the threshold power (within 10%) was observed whereas the effect of the superfluid He around the sphere would be expected to change significantly when approaching the λ transition.

We are therefore confident that the bistable behaviour we observe is due to the Kerr non-linearity of silica. As first pointed out by Braginsky *et al.* [1], once we observe this intrinsic nonlinearity, silica WGM microresonators become well-suited for the realization of sub-shotnoise effects such as QND measurements with cross-Kerr interaction [15] or intensity squeezing in a Kerr bistable device [16].

These experiments at low temperature are also a first step toward the realization of a thresholdless microlaser. We have recently used the unique properties of the WGM resonators to demonstrate a very low threshold laser based on neodymium-doped silica microspheres [17]. In these experiments, the cavity enhancement, described by the Purcell factor η [18], is limited by the homogeneous linewidth $\Delta\nu_{\mathrm{H}}$ of the lasing transition. When $\eta \geq 1$, the spontaneous emission is "forced" into the corresponding cavity mode and the laser should exhibit a thresholdless behaviour [5]. This regime has been studied in multilayer semiconductor microstructures but η was limited in practice to about 1 to 10% [19,20]. In our case, $\Delta\nu_{\mathrm{H}} \approx 1500$ GHz at 300 K, and η is of the order of 4% [17]. Cooling the silica matrix efficiently reduces $\Delta\nu_{\mathrm{H}}$ which is expected to decrease like T^2 down to about 10 K [21]. Operating at liquid-helium temperatures should therefore allow us to reach a thresholdless regime with $\eta > 1$.

Besides, our previous results at 300 K have also shown that only $N_{\mathrm{th}} \approx 10^6$ ions are excited at threshold in the most confined mode with $V_{\mathrm{M}} \approx 300$ μm^3. Since N_{th} is proportional to $\Delta\nu_{\mathrm{H}}$, we also hope to realize a fully-quantum thresholdless laser with only a few excited ions. The quantum fluctuations should then play a major role, as demonstrated by numerical simulations based on the resolution of Heisenberg-Langevin equations [22,23].

With high Q-factors preserved in 50 μm diameter spheres, another route toward strong CQED effects can be investigated. The enhanced field of a WGM can be used to excite a single molecule brought into the evanescent field close to the sphere [24–26]. One very promising system for single-molecule spectroscopy is dibenzoterrylene embedded in a naphtalene host microcrystal, which has all the desired features at low temperature [27]: simple two-level behaviour on the zero-phonon line (ZPL) without triplet leaks, narrow and very photostable lines with high fluorescence efficiencies. The ZPL oscillator strength at $\lambda = 757$ nm corresponds to a radiative lifetime $\tau_{\mathrm{ZPL}} \simeq 60$ ns and the homogeneous width of the transition is of the order of 30 MHz at 1 K. Using $V = 300$ μm^3 for the WGM mode volume, the coupling between the molecule and the vacuum field in the cavity described by the Rabi pulsation is $\omega_{\mathrm{R}} = k[(3/16\pi^2)((\lambda/N)^3/V)(\omega/\tau_{\mathrm{ZPL}})]^{1/2}$ where $k < 1$ accounts for the evanescent wave decrease outside the sphere, and the refractive index mismatch between naphtalene and silica.At a distance of 10 nm from the sphere, $k \simeq 0.3$ and $\omega_{\mathrm{R}}/(2\pi) \simeq 30$ MHz. If high enough Q-factors can be preserved with such micro-crystals on the sphere's surface (like what is observed with a fiber tip [28]), the energy exchange rate between the emitter and an empty single cavity mode will be comparable to the loss rates of the system (molecular dipole decay and cavity losses) and the strong coupling regime of CQED should be reached [4].

To summarize, we have observed very narrow WGM resonances with high Q-factors up to 10^9 on 50 μm to 100 μm in diameter silica microspheres immersed in a superfluid helium bath. We also give evidence for the observation, in these microresonators, of the intrinsic Kerr bistability with a 10 μW threshold. During our very first experiments at 2 K, we have already observed laser action on a spoiled doped sphere with a low $Q \simeq 10^6$. The high Q-values reported here will certainly help us to achieve extremely low thresholds on the Nd^{3+}-microlaser. The very small cavity mode volume combined with the preserved high Q-factor in a cryogenic environment is also favourable for other CQED projects like the study of a single molecule coupled to a WGM, or the use of the Kerr effect to produce a non-linear interaction between neighbouring WGMs.

We acknowledge stimulating discussions with P. Domokos, N. Dubreuil and W. von Klitzing. This research has been supported in part by a France Telecom CNET/CNRS Contract (91.1B 180) and by the CNRS Ultimatech program. Laboratoire Kastler Brossel is a Unité de Recherche de l'École Normale Supérieure et Université Pierre et Marie Curie, associated to the Centre National de la Recherche Scientifique (CNRS).

References

1. V.B. Braginsky, M.L. Gorodetsky, V.S. Ilchenko, Phys. Lett. A **137**, 393 (1989).
2. L. Collot, V. Lefèvre-Seguin, M. Brune, J.-M. Raimond, S. Haroche, Europhys. Lett. **23**, 327 (1993).
3. "Optical Processes in Microcavities", Advances Series in Applied Physics (Vol. 3), edited by R.K. Chang, A.J. Campillo (World Scientific, 1996).
4. S. Haroche, in *Cavity Quantum Electrodynamics*, Proceedings of the Les Houches Summer School of Theoretical Physics, Session LIII, Les Houches, 1990, edited by J. Dalibard, J.-M. Raimond, J. Zinn-Justin (North-Holland, Amsterdam, 1992).
5. Y. Yamamoto, R.E. Slusher, Physics Today **46**, 66 (1993).
6. D.S. Weiss, V. Sandoghdar, J. Hare, V. Lefèvre-Seguin, J.-M. Raimond, S. Haroche, Opt. Lett. **20**, 1835 (1995).
7. M.L. Gorodetsky, A.A. Savchenkov, V.S. Ilchenko, Opt. Lett. **21**, 453 (1996).
8. F. Treussart, J. Hare, L. Collot, V. Lefèvre, D.S. Weiss, V. Sandoghar, J.-M. Raimond, S. Haroche, Opt. Lett. **19**, 1651 (1994).
9. V.S. Ilchenko, M.L. Gorodetsky, Laser Phys. **2**, 1004 (1992).
10. C.E. Wieman, L. Hollberg, Rev. Sci. Instrum. **62**, 1 (1991). We measured using a self-homodyne scheme a laser linewidth of 70 kHz for the 810 nm diode, and 1.5 MHz for the DBR laser.
11. O.V. Lounasmaa, *Experimental principles and Methods below 1 K* (Academic Press, London, 1974).
12. This shift results from the contributions of the index temperature change and the expansion coeficient and one has: $(1/\nu)(\mathrm{d}\nu/\mathrm{d}T) = -(1/N)(\mathrm{d}N/\mathrm{d}T) - (1/a)(\mathrm{d}a/\mathrm{d}T)$. At 300 K, the thermal frequency shift is negative, of the order of -2.5 MHz/mK. Although $(\mathrm{d}N/\mathrm{d}T)$ is not well known at low T, the small positive thermal shift that we have measured at 6 K agrees with the expansion coefficient of silica which is known to vanish and change sign below 20 K.
13. The Kerr index of silica has been measured by various methods: see K.O. Hill, D.C. Johnson, B.S. Kawasaki, R.I. MacDonald, J. Appl. Phys. **49**, 5098 (1978) and references therein; see also for a recent measurement: A.J. Taylor, G. Rodriguez, T.S. Clement, Opt. Lett. **21**, 1812 (1996).
14. M.H. Edwards, Can. J. Phys. **36**, 814 (1958).
15. J.F. Roch, K. Vigneron, Ph. Grelu, A. Sinatra, J.Ph. Poizat, Ph. Grangier, Phys. Rev. Lett. **78**, 634 (1997).
16. A. Lambrecht, T. Coudreau, A.M. Steinberg, E. Giacobino, Europhys. Lett. **36**, 93 (1996).
17. V. Sandoghdar, F. Treussart, J. Hare, V. Lefèvre-Seguin, J.-M. Raimond, S. Haroche, Phys. Rev. A **54**, R1777 (1996).
18. The Purcell factor η is the ratio of the spontaneous emission rate in a single cavity mode of volume V_{M}, to the usual total spontaneous emission rate over all the modes of a bulk medium. As long as $\Delta\nu_{\mathrm{H}}$ is greater than the cavity mode linewidth ν_{L}/Q, one has $\eta = [(4\pi^2/3)(\Delta\nu_{\mathrm{H}}/\nu_{\mathrm{L}})(V_{\mathrm{M}}/\lambda^3)]^{-1}$.
19. A.J. Horowicz, H. Heitmann, Y. Kadota, Y. Yamamoto, ppl. Phys. Lett. **61**, 39 (1992); F.M. Matinaga, A. Karlsson, S. Machida, Y. Yamamoto, T. Suzuki, Y. Kadota, M. Ikeda, Appl. Phys. Lett. **62**, 443 (1993).
20. U. Mohideen, R.E. Slusher, Phys. Rev. Lett. **73**, 1785 (1994).
21. R.M. MacFarlane, R.M. Shelby, J. Lumin. **36**, 179 (1987).
22. L. Davidovich, Rev. Mod. Phys. **68**, 127 (1996).
23. P. Domokos, J.-M. Raimond, S. Haroche, I. Protsenko, L. Davidovich, Acta Phys. Slov. **47**, 273 (1997).
24. W.E. Moerner, L. Kador, Phys. Rev. Lett. **62**, 2535 (1989).
25. M. Orrit, J. Bernard, Phys. Rev. Lett. **65**, 2716 (1990).
26. D.J. Norris, M. Kuwata-Gonokami, W.E. Moerner, Appl. Phys. Lett. **71**, 297 (1997).
27. F. Jelezko, Ph. Tamarat, B. Lounis, M. Orrit, J. Phys. Chem. **100**, 13893 (1996).
28. J.C. Knight, N. Dubreuil, V. Sandoghdar, J. Hare, V. Lefèvre-Seguin, J.-M. Raimond, S. Haroche, Opt. Lett. **20**, 1515 (1995).