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Very low threshold whispering-gallery-mode microsphere laser

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We report on the realization of a whispering-gallery-mode laser based on neodymium-doped silica microspheres. Absorbed pump powers at threshold are as low as 200 nW. The linear variation of the threshold with the loss factor of the cavity mode has also been observed. We discuss the potential of this system as a permanent microlaser operating with a few active ions at liquid-helium temperature. [S1050-2947(96)50309-4]

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Miniature resonators are attractive systems both for fundamental physics research in the field of cavity quantum electrodynamics (CQED) [1] and for use in optoelectronic systems. In multilayer semiconductor microlasers and in microdisks, the coupling of the gain medium can be limited to only a few modes of radiation so that strongly enhanced stimulated emission is achieved and ''thresholdless'' lasers are observed [2]. However, these devices are operated in the low-Q CQED regime. Small Fabry-Pérot cavities with very high finesses have been used both for high-Q CQED experiments and for single-atom laser action, but these experiments involve special supermirrors and atomic beams and remain technically difficult [3]. We have chosen instead to study fused-silica microspheres which act as high-Q small-volume optical resonators owing to the properties of whispering-gallery modes (WGM's). Light in such modes is trapped near the surface by repeated total internal reflections and travels in a great circle around the sphere with virtually no loss except for residual absorption and scattering in the dielectric. With WGM volumes of several hundreds of cubic wavelengths, the electric-field amplitude per photon becomes sufficiently high to realize CQED experiments or to observe nonlinear optical effects with a small number of photons. Whispering-gallery-mode lasers have been demonstrated from the early days of lasers by Garrett, Kaiser, and Bond with millimetric CaF_2:Sm^{2+} spheres [3]. Among the many cavity-enhanced or nonlinear optical effects obtained in microdroplets, lasing has also been reported in ethanol droplets doped with rhodamine [5] and on polystyrene spheres covered with rhodamine [6]. Recently, stimulated Raman scattering has been observed in CS_2 microdroplets with a threshold of three photons only coupled into the pumped WG mode [7]. However, experiments with droplets suffer from their short lifetime since the droplet evaporates and is often in free fall. On solid spheres, laser oscillation has been observed with large neodymium-doped yttrium aluminum garnet spheres (with a diameter of 5 mm) with a threshold of 100 mW [8] and with Nd:glass spheres smaller than 40 μm placed on a plate in the focus of a high power laser [9], but the transparency of the host glass was rather poor and WGM-mode characterization was not possible. By comparison, pure silica microspheres have been demonstrated [10–12] to provide a permanent system that combines relatively small mode volumes with the highest observed Q's ($>2.10^9$) corresponding to photon storage times of the order of 1 μs. We have used microspheres of radius $a \approx 25-50$ μm, formed by heat-fusing the tip of a length of doped silica wire. Such resonators are reproducible and easy to manipulate, and efficient coupling of light in the most confined WGM's can be achieved with a high-index prism, which was not possible in Refs. [5,9]. The low thresholds reported here for laser action in Nd-doped silica microspheres show that it seems feasible, as discussed below, to build a ''thresholdless'' microlaser with only a few active ions, at liquid-helium temperature. Silica microspheres appear, therefore, as promising resonators for future work on CQED with one atom or ion at low temperature strongly coupled to a few photons in a WGM.

Among the various rare-earth ions that can be investigated, neodymium ions provide a favorable four-level laser system [13] that can be pumped on the $^4I_{9/2}$-$^4F_{5/2}$ transition at around 810 nm with a diode laser. The laser transition $^4F_{3/2}$-$^4I_{11/2}$ (in the 1060–1090 nm range) connects a long-lived upper level to a lower level that is depleted by strong phonon relaxation so that population inversion is easily achieved.

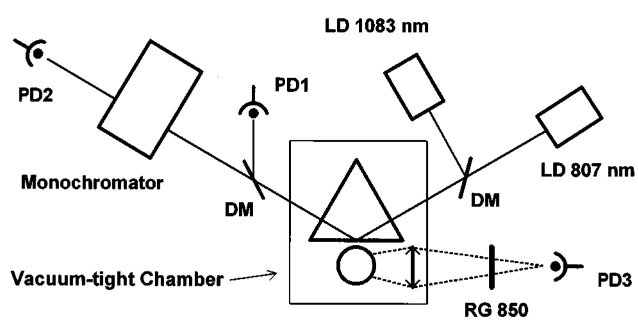

FIG. 1. Experimental setup: WGM's are excited by evanescent-wave coupling with a high index prism across the sphere-prism gap g, which can be adjusted with nanometric precision. The pump beam at 807 nm and the 1080-nm probe beam can be superimposed with a dichroic mirror (DM). The pump absorption is monitored with PD1. The laser light (or fluorescence) is detected at the exit of the monochromator (resolution 0.02 nm) with PD2. The side fluorescence of the doped sphere is measured with photodiode PD3, after elimination of the scattered pump light with the RG850 filter. The microsphere can also be viewed with a CCD camera coupled to a stereomicroscope (not shown here).

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Figure 1 shows the schematics of the experimental setup viewed from the top. The microsphere is made from a fiber with a core of 20 μm in diameter containing about 0.2% Nd in weight. We first remove the fiber cladding by immersion in hydrofluoric acid. The doped wire formed by the exposed core is then melted in our closed-chamber setup using a CO_2 laser focused on the wire's end [11]. Under the influence of surface tension one obtains quasispherical structures with diameters between 50 and 80 μm, which we refer to as microspheres. The wire that remains connected to the sphere serves as a convenient handle for its manipulation without affecting the ring-shaped WGM's located near the sphere's equator [10,11]. We pump the ions using a resonant WGM excited via evanescent wave coupling from a high-index prism. The emission from the sphere is then coupled out via the same prism and is diagnosed. The pump source is a diode laser (Sharp LT017MDO) operating at 807 nm. By adjusting the diode current, resonant excitation of a sphere's WGM can be observed as a dip on the light transmitted to the photodiode PD1. The width of this resonance due to the absorption by the Nd ions varies between 420 and 600 MHz when different spheres are compared. This corresponds to moderate Q factors in the 10^6 range, in agreement with the initial fiber attenuation of 55–80 dB/m, which shows that the fusion process does not alter substantially the Nd concentration. The WGM resonances at the laser transition wavelength are examined independently with a second diode laser at 1083 nm. The absorption in the 1060–1080-nm region remains quite weak so the WGM resonances have much higher intrinsic Q's on the order of several 10^8, possibly limited by a residual thin water layer since higher Q's were observed at 780 nm [11,12]. Light emitted out of WGM's in the fluorescence band and recoupled into the prism is separated from the reflected pump beam and then passed through a monochromator (with a resolution of 0.02 nm) onto photodiode PD2. Another photodiode (PD3) together with a filter (RG850) is used to measure the fluorescence light that is collected by imaging the sphere from the side.

We will discuss here the results obtained on a typical microsphere. Its diameter was 56 μm and its Nd content induced a WGM linewidth of 430 MHz at the pump wavelength (equivalent to an attenuation of 57 dB/m). We first recorded with the monochromator the fluorescence spectrum on the $^4F_{3/2}$-$^4I_{11/2}$ transition (dashed line in Fig. 2). Its broad profile results from the superposition of ten different lines between the Stark sublevels of the upper and lower states, each line overlapping with its neighbors due to their homogeneous and inhomogeneous widths, both of the order of 6 nm [14]. Due to the presence of Al_2O_3 as a codopant (0.5%), the peak wavelength is shifted from about 1080 nm (for silica with a few percent of germanium oxide) down to about 1060 nm.

In general, we observed laser emission in a multimode regime with modes distributed over all this spectral region, as displayed by the solid line in Fig. 2. The lasing modes nicely reproduce the quasiperiodic WGM spectrum where adjacent modes differ by one unit of angular momentum l [15]. Their spacing of 4.5 nm at 1083 nm agrees with the equivalent "free spectral range" of the sphere ($c/2\pi Na$) deduced from the sphere's diameter $2a$ and the refractive index $N=1.45$. On closer examination each of the peaks in

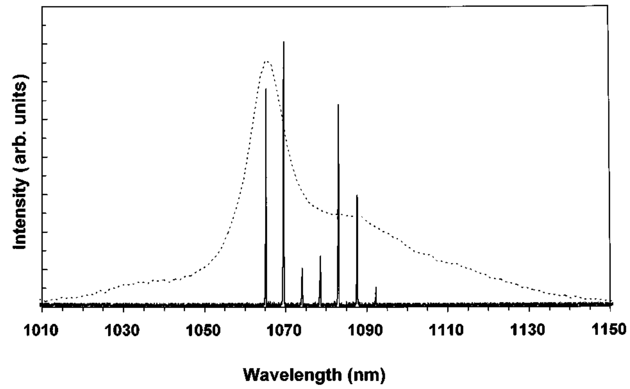

FIG. 2. Typical multimode laser spectrum (full line) from a microsphere of diameter 56 μm doped with about 0.2% Nd in weight. The dashed line is the fluorescence spectrum (not to scale) recorded below threshold. The equally spaced laser modes give the "free-spectral range" of the microsphere.

Fig. 2 contains three or four neighboring laser lines with a separation of about 0.06 nm. These modes correspond to different m azimuthal numbers $|m|=l,l-1,l-2...$with increasing spatial extensions in the polar direction. Similar to the transverse modes of a nearly confocal Fabry-Pérot, they would be degenerate for a perfect sphere. From their spacing of 0.06 nm, we estimate the ellipticity of this "microsphere" to be 1% [16]. The occurrence of modes with $|m|\neq l$ is consistent with the rather large polar extension of the pump mode, observed with the charge-coupled-device (CCD) camera. In principle, there are other "transverse" WGM's labeled by a radial order number n and related to the radial penetration depth of the field. We did not see any evidence of these modes. The equally spaced laser lines suggest that the pump mode has the minimum radial extent ($n=1$), which efficiently selects $n=1$ lasing modes. The spectrum discussed here corresponds to laser light propagating counterclockwise in the sphere (positive m values). As expected, we also observed laser light generated in the clockwise direction ($-m$), which doubles the number of lasing modes sustained at the same time in the microsphere.

Another experimental feature concerns the polarizations of the pump and laser modes. TE resonances (with the electric field nearly perpendicular to the equatorial plane) have a penetration depth outside the sphere slightly greater than TM modes (with the electric field nearly radial) associated with the frequency shift between the nearly identical TM and TE spectra [15]. This effect amounts to an effective gap g slightly smaller for TE modes, which makes optical tunneling of the pump light more efficient for TE modes while TM modes preserve better Q's. This is our interpretation explaining why we always obtain laser action with TE pump modes while the laser modes are TM polarized.

We measured the microlaser threshold by varying the incident pump power and monitoring the intensity of one of the longitudinal modes, using the monochromator as a filter. The power recorded on PD2 was then plotted as a function of the power absorbed from the pump as measured from the dip signal on PD1. Figure 3 displays one of the lowest observed thresholds corresponding to an absorbed pump power of 200 nW. It was obtained for a cold-cavity Q of 2×10^8 and for an incident pump power focused on the prism face of about 1

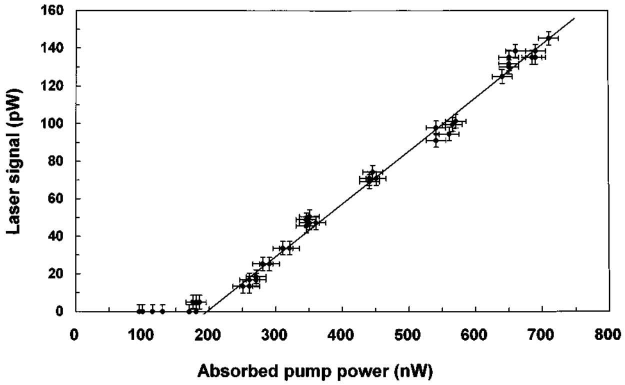

FIG. 3. Intensity of the laser signal recorded on PD2 as a function of the absorbed pump power for the same sphere as above in the multimode regime. The cold-cavity Q was 2×10^8. The threshold is well marked at 200 nW.

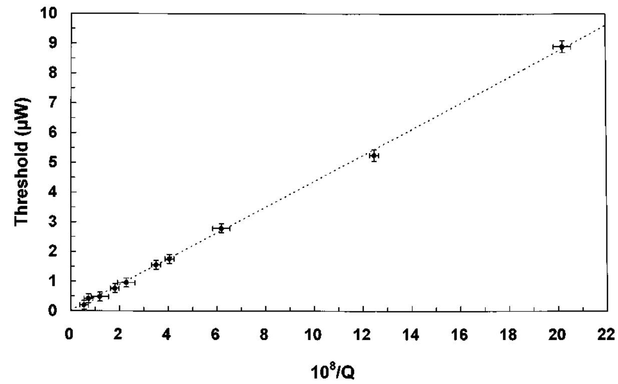

FIG. 5. Threshold power as a function of the loss factor Q^{-1} of the cavity showing the expected linear variation.

mW. The coupling efficiency of the laser emission into the prism was about 5%. Extra losses along the detection path (about 90%) result in a measured laser output between 10 and 150 pW, as shown in Fig. 3. The fluorescence side light recorded simultaneously on PD3 is shown in Fig. 4. The same threshold is clearly visible by the marked change of slope above 200 nW of absorbed pump power, signaling the expected clamping of the population inversion at its threshold value. This sudden slope change indicates that all the lasing modes share the same threshold within experimental precision. The residual slope observed in Fig. 4 above 200 nW absorbed power means that about 10% of the excited ions do not participate in any laser line.

The threshold for laser action is expected to increase linearly with the losses of the cavity. To check this law, we approached the prism to the sphere, therefore increasing the WGM's linewidth and the cold-cavity loss factor Q^{-1}. For each position, we measured the threshold and obtained the linear variation as a function of Q^{-1} shown in Fig. 5.

We also looked for laser lines in the 900–940-nm fluorescence band ($^4F_{3/2}$-$^4I_{9/2}$ transition). Quasiperiodic multimode emission was obtained simultaneously with two other lines at around 1060 nm. For smaller sphere-prism gaps, we even observed that lasing was suppressed at 1060 nm while a few modes were still present around 930 nm. This is a consequence of the wavelength-dependent sphere-prism coupling efficiency. Since it varies roughly as $\exp(-4\pi g/\lambda)$, short-wavelength lasing modes are less broadened by the presence of the prism. However, since we had no means to measure the cold-cavity Q factor at this wavelength, we did not investigate this domain in more detail.

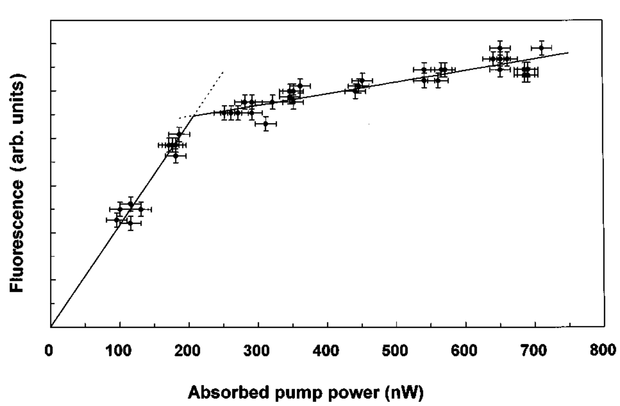

FIG. 4. Side-fluorescence intensity as a function of the absorbed pump power recorded simultaneously with the laser signal shown in Fig. 3.

The lowest threshold reported here can be compared to the expectation from a basic four-level system model. Adapting textbook formulas to our resonators [13], we can write the population-inversion density at threshold as $\Delta n_{\mathrm{th}}=(4\pi^2/3)(\Delta\nu_h/\nu_L)(\tau_{\mathrm{sp}}/\tau_{\mathrm{cav}})\lambda^{-3}$, where the homogeneous linewidth of the laser transition at ν_L is $\Delta\nu_h\approx1500$ GHz at room temperature, $\tau_{\mathrm{cav}}\approx100$ ns the decay time of energy in the sphere for a Q factor of 2×10^8, and $\tau_{\mathrm{sp}}\approx1$ ms the radiative lifetime of the upper level in free space. We get a density $\Delta n_{\mathrm{th}}\approx600$ excited ions/μm^3 for a single laser mode. Let us emphasize that this density corresponds to only 2×10^5 ions in the most confined modes available in such a sphere. We then infer the power to be absorbed from the pump mode $P_{\mathrm{th}}=\Delta n_{\mathrm{th}}V_P(h\nu_P/\tau_2)\alpha^{-1}$, where V_P is the pump-mode volume, $\tau_2\approx0.5$ ms is the total decay rate of the upper level of the lasing transition, and $\alpha\approx0.5$ is the estimated efficiency of the pumping mechanism toward this level [17]. By a rough measurement of the luminescent band around the sphere's equator, we estimate the pump-mode volume to be of the order of 3000 μm^3, which is about ten times the minimum mode volume available in the sphere discussed here. If there were only one laser mode, the pump power absorbed at threshold would be $P_{\mathrm{th}}\approx2$ nW. From the spectrum of the lasing modes described above, we estimate that the pump mode is able to sustain 60 to 80 lasing modes at the same time. This explains the observed 200 nW quite well.

The thresholds reported here are smaller than for conventional Nd-doped fiber lasers [18] by more than three orders of magnitude. This is mostly due to the very low losses of WGM's. The threshold can be further reduced at room temperature by decreasing the number of modes lasing simultaneously. First of all, we plan to apply the mode identification methods that we have developed using a fiber tip as a near-field probe [19] in order to select a pump mode with the maximal confinement (i.e., $n=1$ and $m=l$) and therefore eliminate the $|m|\neq l$ lasing modes. Second, we also wish to tune the WGM spectrum of a given sphere to match the gain profile of the ions. By comparing different spheres, we have already observed that the number of lasing modes seems to depend on slight changes of the sphere's radius that modify the free spectral range and the position of the WGM resonances. In particular, although the broad multilevel structure of the laser transition is favorable to multimode emission, we have obtained in some circumstances single-mode operation. Mechanical tuning by the application of strain with piezoelectric transducers could give us an elegant way to control the single-mode regime [20]. We have used a self-heterodyning setup with a 11-km-long fiber to measure the laser linewidth. It was found to be about 65 kHz in the multimode regime, but this measurement still needs to be repeated under single-mode operation. If lower linewidths are obtained, interesting applications could be found when coupling lasing microspheres to fibers or planar waveguides. We also plan to investigate other rare-earth ions: upconversion in erbium, for instance, could produce green laser light at 550 nm with an 800-nm diode laser as a pump.

A further major step will be to operate at liquid-helium temperature in order to lower the homogeneous linewidth $\Delta\nu_h$ of the Nd^{3+} transitions [21]. Cavity enhancement is often described by the Purcell factor η given in our case by $[(4\pi^2/3)(\Delta\nu_h/\nu_L)(V_m/\lambda^3)]^{-1}$ for a mode of volume V_m, and lasing without threshold is observed when $\eta \geqslant 1$ [2]. Since $\Delta\nu_h$ decreases roughly like T^2, η, which is limited to about 4% at room temperature, will become greater than 1. We would then be able to achieve a ''thresholdless'' microlaser in which the active medium would be made of a few ions only. Due to the large inhomogeneous broadening $\Delta\nu_{\text{inh}}$, a small fraction $(\Delta\nu_h/\Delta\nu_{\text{inh}})$ of the ions will be selectively coupled to the cavity mode and the Nd doping content should be chosen accordingly. The upper Stark sublevel of the $^4F_{3/2}$ state will also be empty at these low temperatures so that several unwanted laser lines will be suppressed. The very low laser thresholds reported here are also promising for cavity-QED experiments on the transition from the $^4F_{3/2}$ to the $^4I_{9/2}$ ground state. Since $\Delta\nu_h \approx 2$ MHz has been observed at 1 K on this transition [22], the strong-coupling regime could be achieved between a few neodymium ions and a few hundred photons in a WGM.

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[1] 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, and J. Zinn-Justin (North-Holland, Amsterdam, 1992) and references therein.
[2] Y. Yamamoto and R. E. Slusher, Phys. Today **46**(6), 66 (1993).
[3] R. J. Thompson, G. Rempe, and H. J. Kimble, Phys. Rev. Lett. **68**, 1132 (1992); K. An, J. J. Childs, R. R. Dasari, and M. S. Feld, *ibid.* **73**, 3375 (1994).
[4] C. G. B. Garrett, W. Kaiser, and W. L. Bond, Phys. Rev. **124**, 1807 (1961).
[5] H.-M. Tzeng, K. F. Wall, M. B. Long, and R. K. Chang, Opt. Lett. **9**, 499 (1984).
[6] M. Kuwata-Gonokami, K. Takeda, H. Yasuda, and K. Ema, Jpn. J. Appl. Phys. **31**, L99 (1992).
[7] H.-B. Lin and A. J. Campillo, Phys. Rev. Lett. **73**, 2440 (1994).
[8] T. Baer, Opt. Lett. **12**, 392 (1987).
[9] Y. Z. Wang, B. L. Lu, Y. Q. Li, and Y. S. Liu, Opt. Lett. **20**, 770 (1995).
[10] V. B. Braginsky, M. L. Gorodetsky, and V. S. Ilchenko, Phys. Lett. A **137**, 393 (1989).
[11] L. Collot, V. Lefèvre-Seguin, B. Brune, J. M. Raimond, and S. Haroche, Europhys. Lett. **23**, 327 (1993); D. S. Weiss, V. Sandoghdar, J. Hare, V. Lefèvre-Seguin, J.-M. Raimond, and S. Haroche, Opt. Lett. **20**, 1835 (1995).
[12] M. L. Gorodetsky, A. A. Savchenkov, and V. S. Ilchenko, Opt. Lett. **21**, 453 (1996).
[13] A. E. Siegman, *Lasers* (University Science Books, Mill Valley, CA, 1986).
[14] M. M. Mann and L. G. DeShazer, J. Appl. Phys. **41**, 2951 (1970).
[15] S. Schiller and R. L. Byer, Opt. Lett. **16**, 1138 (1991).
[16] H. M. Lai, C. C. Lam, P. T. Leung, and K. Young, J. Opt. Soc. Am. B **8**, 1962 (1991).
[17] M. Shimizu, H. Suda, and M. Horiguchi, Electron. Lett. **23**, 768 (1987).
[18] R. J. Mears, L. Reekie, S. B. Poole, and D. N. Payne, Electron. Lett. **21**, 738 (1985).
[19] J. C. Knight, N. Dubreuil, V. Sandoghdar, J. Hare, V. Lefèvre-Seguin, J. M. Raimond, and S. Haroche, Opt. Lett. **20**, 1515 (1995); **21**, 698 (1996).
[20] A. L. Huston and J. D. Eversole, Opt. Lett. **18**, 1104 (1993); V. S. Ilchenko (private communication).
[21] R. M. Macfarlane and R. M. Shelby, J. Lumin. **36**, 179 (1987).
[22] M. M. Broer, B. Golding, W. H. Haemmerle, and J. R. Simpson, Phys. Rev. B **33**, 4160 (1986).