Available online at www.sciencedirect.com
ScienceDirect
Optics Communications 265 (2006) 33–38
OPTICS COMMUNICATIONS
www.elsevier.com/locate/optcom

Ultra high Q crystalline microcavities

Ivan S. Grudinin *, Andrey B. Matsko, Anatoliy A. Savchenkov, Dmitry Strekalov, Vladimir S. Ilchenko, Lute Maleki

Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA 91109, United States

Received 28 February 2006; accepted 2 March 2006

Abstract

We report on the fabrication technique of ultra high Q optical crystalline whispering gallery mode microresonators and discuss their properties. The technique is suitable for the majority of available optical crystals and for production of resonators with small size. To validate the method, we made CaF_2 resonators with Q factors exceeding 4×10^8 and a diameter smaller than 100 μm. A single mode resonator has also been fabricated. Possible utilization of these new resonators in quantum optics is discussed.

PACS: 42.50.−p; 42.60.Da

Keywords: Whispering gallery modes; Microcavity; Single mode resonator; Diamond turning; Cavity QED

1. Introduction

Although the whispering gallery mode resonators (WGMR) were invented almost a century ago, significant advances in their actual utilization appeared only in the last couple of decades following the invention of efficient resonator-light coupling methods. The WGM resonances, also known as morphology dependent resonances, have proven to be useful in many photonics and optics applications. Miniaturization of these resonators makes new applications possible in precision physics experiment [1,2].

Historically, the WGM resonators have been fabricated with a variety of materials including silicon [3], sapphire, fused silica [4], calcium fluoride, lithium niobate [5] and other optical glasses and crystals. The optically transparent crystals are particularly interesting for fabrication of the resonators because they naturally have many specific optical [6] and spectroscopic features, such as zero phonon lines, which are absent in amorphous materials. Another advantage of crystalline materials for the fabrication of the resonators is that they can be stable against humidity, as in the case of CaF_2. Quality (Q) factor of the fused silica microspheres and microdisks degrades when exposed to the atmospheric water vapor [7]. The resonators made with a non-hygroscopic crystal are free of this disadvantage.

The high Q factor of the resonators and their small size (small volume of the WGMs) is of great importance for a number of applications. The record Q factor of 2×10^{10} was demonstrated in CaF_2 resonators in our group at laser wavelength of 1550 nm [8]. However, the resonators were rather large, about 10 mm in diameter. We have recently improved this record to $Q=5.3\times10^{10}$ for 5 mm and $Q=4\times10^{10}$ for 1.5 mm resonators measured at wavelength of $\lambda=1064$ nm. The goal of the present work is to show the possibility of fabrication of ultra high Q crystalline microresonators and highlight their possible applications.

We report on the development of a fabrication technique for the crystalline microresonators. The technique is based on a home-made diamond turning apparatus. The quality of the surface of the resonators produced with

* Corresponding author. Tel.: +1 818 393 7090; fax: +1 818 393 6773.
E-mail addresses: grudinin@caltech.edu, grudinin@jpl.nasa.gov (I.S. Grudinin).
URL: http://horology.jpl.nasa.gov/quantum/.

0030-4018/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.optcom.2006.03.028

this technique is characterized by less than 0.2 nm surface roughness. The quality factor of the microresonators fabricated with CaF_2 exceeds 10^8 and is stable in normal atmospheric conditions. We discuss the potential of these resonators for creating breakthroughs in quantum optics and single ion spectroscopy.

2. Fabrication of new microresonators

In this section, we discuss the fabrication technique of optical crystalline WGM microresonators. The diameter of resonators can be as small as a few tens of microns and their geometrical features, such as surface curvature or profile, can be precisely engineered. We have fabricated magnesium fluoride and calcium fluoride resonators to validate the technique. Some of these cavities are shown in Figs. 1–3.

The fabrication process includes two steps. The first step is the diamond turning process, which employs computer control of a precision lathe. The structures obtained at this step are engineered to about 40 nm precision and have optical Q factors of up to 10^7. If higher Q factors are needed, additional optical polishing has to be performed. This polishing step naturally modifies the structure that was initially obtained by the diamond turning process.

Special diamond turning regimes should be employed to avoid brittle machining and to achieve a required smoothness of the surface. Such parameters as speed of rotation of the workpiece, feed and cutting angle, diamond cutter geometry and sharpness as well as lubricant are very important in achieving ductile regime of machining [9]. Special attention should be paid to vibration isolation of the turning process. In our diamond turning setup we use a homemade air bearing to provide the required stiffness and repeatability of workpiece rotations. Commercial brushless motor and a simple magnetic clutch were used to rotate the bearing. The optical polishing is performed by application of polycrystalline diamond abrasives. Polishing is performed in several steps with decreasing diamond grit sizes followed by the cleaning process. It was found that small particles are extremely difficult to remove from the surface of the material. The cleaning process is the most critical in achieving ultrahigh Q factor. Cleaning was performed in a clean room environment with use of organic solvents.

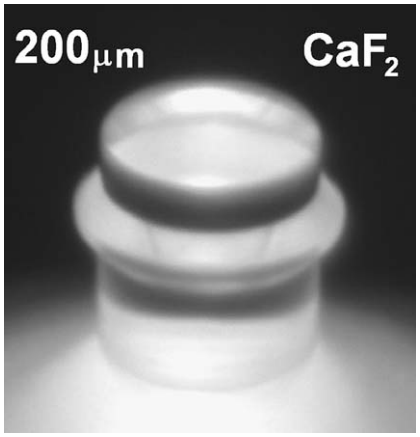

Fig. 1. Calcium fluoride microresonator.

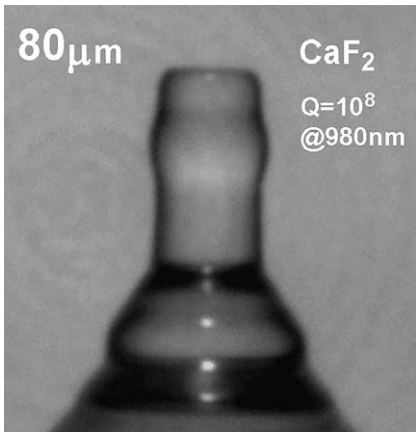

Fig. 2. Calcium fluoride microresonator 80 μm in diameter, $Q = 10^8$.

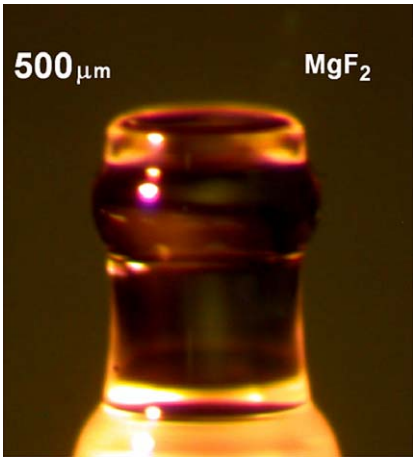

Fig. 3. Magnesium fluoride resonator.

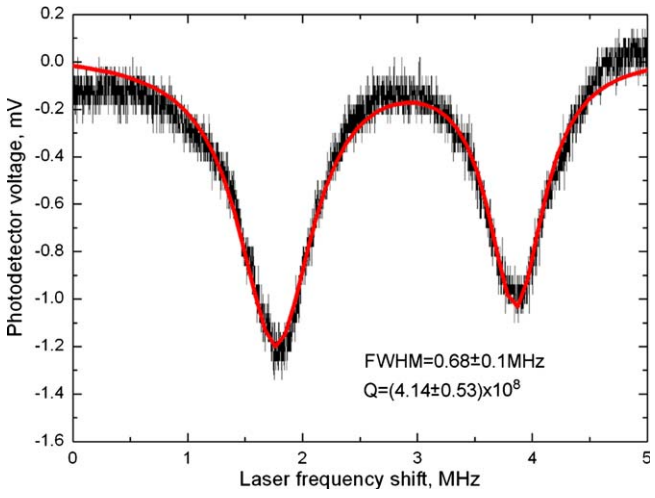

Fig. 4. Resonant curve with Lorentzian fit for CaF_2 resonator 100 μm in diameter. Rayleigh scattering induced splitting is present.

The optical Q factors of the resonators after polishing are limited by contributions from four main sources: scattering by the dust particles accumulated on the resonator surface, absorption in the resonator material, radiation leakage, and surface scattering. We assume that the optical power circulating inside the resonator is below the threshold of nonlinear effects, such as Raman lasing or thermo-optical instability [10]. In a separate report, we will show that threshold for Raman lasing in fluoride cavities is almost two orders of magnitude lower than in fused silica cavities with similar size and Q factor.

Scattering by the dust particles would be the main restriction on the quality factor if a clean room environment is not provided. Once inside the clean room environment or vacuum, the Q factor can be preserved on a very high level for indefinite lengths of time. Aside from dust accumulation, optical polishing leaves some amount of nanoparticles embedded into the resonator surface.

It is known that water is detrimental for the Q factor of fused silica devices due to formation of irregular adsorption layers [11]. Fortunately, such crystals as CaF_2 and MgF_2 have chemical properties that reduce accumulation of water on the surface. Diffusion of water into crystalline lattice is inhibited, while water monolayer (a one molecule thick layer) is always present on the surface.

The intrinsic absorption of the resonator material is the second important effect restricting the value of the measured Q factor. The absorption is a function of crystal growing process and is defined by the optical properties of a crystal at a given wavelength. This parameter is particularly small in crystalline materials, since they can be produced with high purity. Recent developments in UV lithography have stimulated the production of extremely pure CaF_2 monocrystals. However, even in those purest crystals the 'reference-sheet' absorption rarely falls below 10 ppm/cm, which corresponds to quality factors on the order of 10^{10}. Unfortunately, not much data are available for the optical absorption coefficients, since there is no technique capable of measuring this coefficient with high precision when absorption becomes very weak. Interestingly, the WGM resonators may be used to measure internal absorption with good precision if the Q factor is only limited by internal absorption. For example, the absorption coefficient reported by crystal producer is $\alpha = 2 \times 10^{-5}\,\mathrm{cm}^{-1}$. According to the expression for the internal absorption limited Q factor $Q = 2\pi n/(\alpha\lambda)$, this corresponds to $Q = 4.2 \times 10^9$ at wavelength of $\lambda = 1064$ nm. Here, n is the refractive index of material. On the other hand, with this piece of crystal we have measured $Q = 5.3 \times 10^{10}$, which gives more accurate upper estimate of absorption coefficient $\alpha = 1.6 \times 10^{-6}\,\mathrm{cm}^{-1}$.

The radiation leakage is significant only for the resonators with extremely small diameters not exceeding several micrometers, which is not the case in our study.

Finally, the surface roughness is made small by our polishing technique, as confirmed by measurements with an atomic force microscope (AFM). In its contribution to surface scattering factor, the surface roughness is accompanied by the subsurface material damage. This damage is represented by the microfractures and embedded particles and is usually confined within a shallow surface layer of the material. The thickness of this layer is determined by the polishing and grinding process. AFM studies have shown the small amount of diamond particles embedded into the resonator surface. This could be useful in quantum optics, since quantum dot containing diamonds or other nanoparticles can be embedded this way to create a coupled system without noticeable degradation of Q factor.

We have fabricated a calcium fluoride resonator with 100 μm diameter and optical Q factor of 4×10^8 (Fig. 4). The surface roughness does not restrict the value of the quality factor. The measurements performed with AFM showed that the surface roughness of small resonators is about the same as for the large disks with Q factor of 10^9 and is practically negligible (Fig. 5). Let us estimate the maximum Q factor that could be achieved in the resonators with a measured surface roughness. We will use the expression derived in [12]

$$Q \approx \frac{3\lambda^3 a}{8n\pi^2 B^2 \sigma^2}, \tag{1}$$

where λ is the wavelength, a is the radius of the resonator, n is the refraction index of the resonator material, B is the correlation length, and σ is the roughness. Let us assume that $\lambda = 1$ μm, $a = 50$ μm, $n = 1.43$, $B = 2$ nm, and $\sigma = 0.33$ nm. For this set of parameters, we obtain $Q = 4 \times 10^{12}$. The estimated value of the Rayleigh scattering limited Q factor scales proportionally to the radius and is still much higher than any Q factor observed in WGM resonators.

The resonator was fabricated with vacuum UV grade CaF_2 and its Q factor was limited by residual contamination of the surface with polishing agents.

It was found that when an excimer grade CaF_2 is used, it is possible to achieve a Q factor higher than in any other

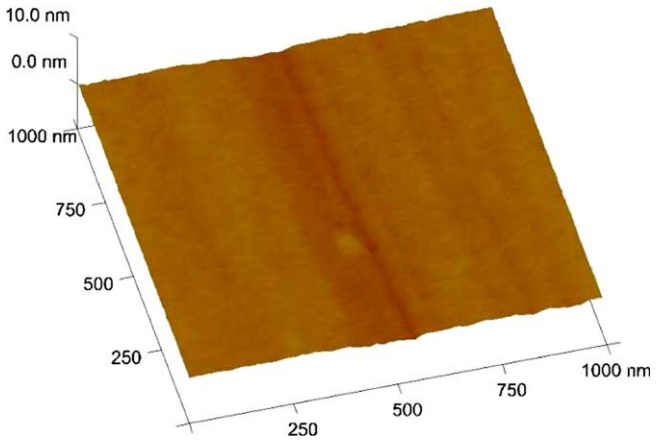

Fig. 5. AFM surface scan of the large 1.6 mm in diameter CaF_2 resonator with Q factor of 10^9. Some polishing groves are visible. Surface roughness is 0.326 nm rms.

open resonator and any other WGM resonator. The 5.5 mm in diameter CaF_2 WGM resonator was fabricated with Q factor of $(5.31 \pm 0.04) \times 10^{10}$ measured with a Nd:YAG laser at wavelength of 1064 nm. In microresonators, the role of Rayleigh and surface scattering becomes more important so the Q factor is lower for smaller cavities [12].

3. Controlling the modal structure of WGMR: single mode resonator

Practically all WGM resonators have more or less complicated modal structures, in other words they have more than one mode per free spectral range. This is an obstacle for many practical applications. The nature of the dense spectrum can be easily understood. The special functions that describe WGM in a perfect dielectric sphere are degenerate. In other words, many modes have the same eigenfrequency. This degeneracy is lifted when the geometry of the resonator is changed to anything different from a perfect sphere. Generally speaking, the special functions should also be different and in most cases it is impossible to resolve the electromagnetic field into any possible kind of special functions. Only approximate analytical and numerical methods may be helpful [13]. The modes that have identical eigenfrequency in a spherical resonator would have different eigenfrequencies in a non-spherical one. This creates a rich optical spectrum in real resonators which are always non-spherical. However, the same degeneracy lifting can be useful, and by engineering the resonator's shape in a special way one could rarify its spectrum. Our fabrication technique allows designing the geometry of the resonator and controlling the structure of its optical spectrum. To demonstrate the versatility of the technique, a single mode calcium fluoride resonator was fabricated, which has only one mode of each polarization per free spectral range (FSR) (see Fig. 6).

Its spectrum is shown in Fig. 8. This particular 5 mm resonator has a loaded Q factor of 8×10^6 limited by occasional dust particles and residual roughness of the surface as well as by the coupling losses. The profile of the resonator is nearly Gaussian as shown in Fig. 7. The Q factor can be as high as 4×10^{10} in a 1.5 mm and 5×10^{10} in a 5 mm resonator as was recently demonstrated in our lab. This could lead to a device with finesse higher than 6×10^6 for a 1.5 mm single mode cavity.

Fig. 6. SEM photograph of a CaF_2 single mode resonator. Light guiding ridge protrudes only about 2.5 μm above the cylinder.

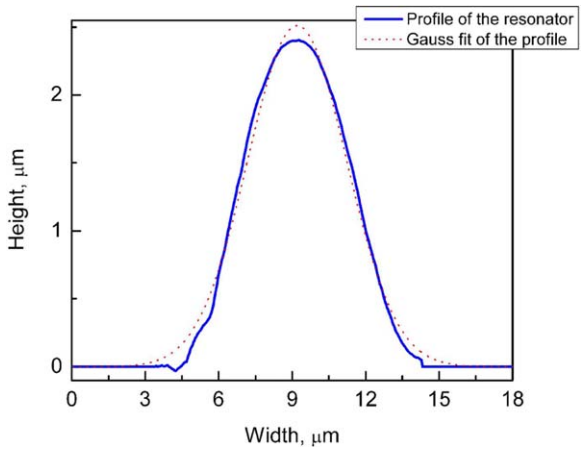

Fig. 7. Nearly Gaussian profile of the single mode resonator shown on Fig. 6.

The spectrum shown was obtained with two angle-polished fiber couplers, one for the input and one for the output. The fiber couplers were made with standard single mode fiber with cladding diameter of about 150 μm and core of about 5 μm. The photodetector signal represents transmission of the resonator. The two families of modes shifted by a few gigahertz were observed, one for each polarization. One of the mode families is shown in Fig. 8. Logarithmic detector measurements have shown that besides the two mode families there were no other modes down to −30 dB level. It should be noted that, given the geometrical parameters of the resonator and the couplers, the single mode regime of this resonator is not a function of coupling but an intrinsic property. Moreover, the logarithmic measurements were performed in a loaded configuration, when the couplers are in contact with the resonator.

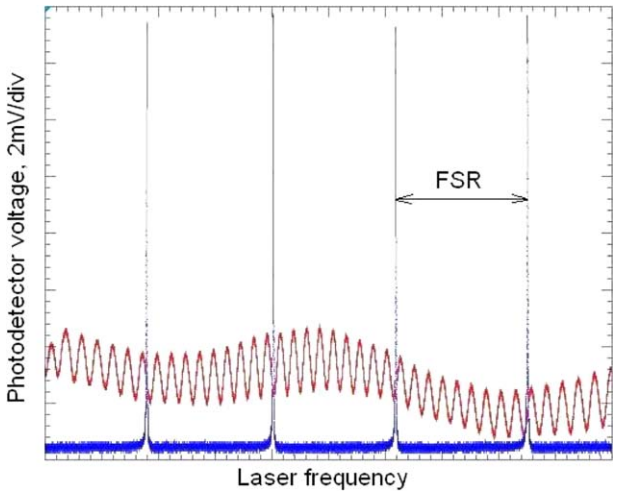

Fig. 8. Single mode WGMR has only one mode of each polarization per free spectral range. 1.3 GHz Fabri–Perot resonator fringes are shown for frequency reference.

This kind of resonator could in principle be fabricated with lithium niobate, whose refractive index can be controlled by applying an electric field. Such a resonator could be used for unique optical measurements and filtering applications. We have also found that crystalline resonator can be easily tuned over a few FSR by applying pressure from a piezo element with efficiency of over 1 GHz per volt (for 500 μm MgF_2 resonator).

It is reasonable to note that such a resonator is no longer a pure WGMR, but rather a hybrid resonator incorporating the features of an optical planar waveguide, or a ridge waveguide, and a conventional WGM resonator. Similar structures have already been investigated [14–16]. Our structure is topologically different and the WGM feature is critical in achieving ultra high optical Q factor. The analysis of this structure is based on the similarity with a single mode gradient waveguide [17,18]. The resonator's profile serves as a confinement structure, in analogy to the refractive index gradient in the single mode optical fiber.

4. Single ion spectroscopy and quantum optics

Ultra high Q crystalline microresonators with small mode volumes are promising for applications in quantum cavity electrodynamics and quantum optics. For instance, the resonators could be used to demonstrate the strong coupling between a WGM and an atom, or to modify the spontaneous emission of an atom interacting with the WGM. Various quantum optics experiments become possible with the new microresonators as the combination of high Q and small mode volume make it easier to utilize optical nonlinearities of the crystalline material. In this section we consider spectroscopy of a single ion in a doped crystalline microresonator as yet another possible application.

The dopants in crystalline WGMR are influenced by the host material of the resonator and by the WGM field structure. The absorption profile of each individual doping ion is broadened homogeneously through interaction with lattice phonons, and shifted through the influence of the lattice itself. This inhomogeneous shift is caused by the slight differences in electronic configurations at the various lattice sites, which create different electric fields for each individual doping ion. The observed inhomogeneous broadening of ions in a crystal host is caused by these shifts. The spectrum of the doping in the crystal is formed as a combination of many overlapping spectra of single ions.

In some crystals the absorption lines of atomic impurities get narrower with decreasing temperature. At liquid helium temperatures the phonon-originated homogeneous broadening fades, leaving the inhomogeneously broadened impurity absorption line with greatly increased intensity. These so-called zero phonon lines make it possible to optically detect single impurity molecules or ions in such crystals [19] using the single molecule spectroscopy techniques. The application of the WGM crystalline resonators may be fruitful for this kind of single ion spectroscopy. Rare earth ions are convenient for observation of the effect because they have many zero phonon line transitions, and relatively strong associated dipole moments for selected transitions. For example, a Sm^{2+} ion in CaF_2 has a dipole moment of 0.63 D for $\lambda = 709$ nm transition [20], which is comparable to organic molecules of pentacene in p-terphenyl matrix, 0.7 D at $\lambda = 592$ nm [21]. Debye is a non-system unit of electric dipole moment. It is equal to the electric dipole moment created by the two charges of 10^{-10} Franklin (or esu) separated by 10^{-10} m ($1\ \mathrm{D} = 10^{-18}$ Fr cm $= 3.33564 \times 10^{-30}$ C m).

The WGM can be used to observe the absorption profile of a single ion if a small enough resonator is doped with rare earth ions in low concentration. At low temperatures the absorption profile of the dopant ions transforms into a "forest" of absorption profiles, corresponding to the individual ions. If the modes of the resonator are scanned over the absorption profiles of the ions, one may expect to observe the interaction of individual photons in a WGM and an ion resulting in quantum effects such as Rabi splitting [22].

The frequency of a WGM can be shifted by use of nonlinear properties of the resonator material (e.g. electro-optical effect in lithium niobate) or by a change of the resonator's geometry. We have performed an experiment for tuning the WG modes by mechanical pressure. The 500 μm resonator made with MgF_2 was installed on a metal support and a piezo element was fixed on top of the resonator in a pre-stressed configuration. By adjusting the piezo voltage we were able to reversibly shift the WGM frequencies by as much as 100 GHz with a sensitivity of 1 GHz per volt.

Let us estimate the efficiency of coupling of a single ion and a WGM for the resonators we have made. Consider a CaF_2 resonator with diameter of 100 μm and optical Q factor of 4×10^8. Assume that it is doped with rare earth ions that have an electric dipole transition moment of $\mu = 1$ D at a wavelength of 1 μm ($\nu = 3 \times 10^{14}$ Hz, $\omega = 2\pi\nu$). The CaF_2 refractive index at this wavelength is $n = 1.43$. Let us assume that we have only one such ion inside the volume occupied by the WGM and that it is located at the point of maximum of electric field. The single photon Rabi frequency and a strong coupling condition may be written as follows:

$$\Omega_r = \frac{\mu E}{\hbar}, \quad \Omega_r \gg (\gamma_{\mathrm{cavity}}, \gamma_{\mathrm{rad}}). \tag{2}$$

Here γ_{cavity} and γ_{rad} are the cavity and the radiative decay rates, respectively; E is the electric field amplitude (CGS units) resulting from a single photon stored in the WGM [23]

$$E = \sqrt{\frac{2\pi\hbar\omega}{V_{\mathrm{eff}} n^2}}, \tag{3}$$

where the effective volume of the mode V_{eff} can be derived using the expression [24]

$$V_{\text{eff}} \cong 3.4\pi^{\frac{3}{2}}\left(\frac{\lambda}{2\pi n}\right)^3 l^{\frac{11}{6}}\sqrt{2(l-m)+1}, \quad l=\frac{2\pi an}{\lambda}. \tag{4}$$

Here l is the radial index of the WGM, a is the radius of the resonator and m is the angular index of the mode. This formula for the volume is derived for silica microspheres and is assumed to be appropriate for our case as the shape of an active region of our resonator is close to spherical. For the given parameters of the resonator we have $l = m = 422$ and $V_{\text{eff}} = 2\times10^{-9}\,\text{cm}^3$. The single photon Rabi frequency is then $\Omega_r/2\pi = 8$ MHz, which is significantly larger than the radiative decay rate $\gamma_{\text{rad}} = 71$ kHz and the cavity decay rate $\gamma_{\text{cavity}} = 0.7$ MHz. Thus the strong coupling condition (2) can be realized. The radiative decay rate of an ion embedded in a lossless dielectric and the cavity decay rate were calculated using the following expressions [23]

$$2\pi\gamma_{\text{rad}} = \frac{4n\mu^2\omega^3}{3\hbar c^3}, \quad 2\pi\gamma_{\text{cavity}} = \frac{\omega}{Q}. \tag{5}$$

5. Conclusion

We have reported on a fabrication technique for crystalline ultra high Q microresonators. Our technique allows precision engineering of the resonator's geometry, which in turn defines its optical spectrum. We have demonstrated CaF_2 resonators with less than 100 μm in diameter that have Q factors exceeding 10^8. These parameters are good enough for the realization of strong coupling of a single photon in a WGM and a single dopant ion. The idea of coupling a WGM cavity mode to optical transitions of ions is not new by itself. The novelty of our idea is in a way the ions are coupled to WGM and in potential advantages a combination of zero phonon lines and crystalline WGM cavities may bring. Taking into account the possibility of making smaller resonators with higher Q factors and a wide range of available inorganic dopants, we see that crystalline WGM resonators open new horizons in quantum optics and atomic physics.

Acknowledgements

The research has been supported by DARPA AOSP Program, and performed at the Jet Propulsion Laboratory, California Institute of Technology, under a contract with NASA. The authors thank Y. Lin and M. Anderson for making the atomic force microscope images of the resonator surfaces. Fabrication and development of crystalline microresonators is a part of I. Grudinin's PhD project.

References

[1] S.M. Spillane, T.J. Kippenberg, K.J. Vahala, Nature 415 (2002) 621.
[2] M.L. Gorodetsky, I. S Grudinin, J. Opt. Soc. Am. B 21 (2004) 697.
[3] M. Borselli, T.J. Johnson, O. Painter, Opt. Exp. 13 (2005) 1515.
[4] D.K. Armani, T.J. Kippenberg, S.M. Spillane, K.J. Vahala, Nature 421 (2003) 925.
[5] A.A. Savchenkov, V.S. Ilchenko, A.B. Matsko, IEEE Photon. Technol. Lett. 17 (2005) 136.
[6] V.S. Ilchenko, A.A. Savchenkov, A.B. Matsko, Phys. Rev. Let. 92 (2004) 043903.
[7] D.W. Vernooy, V.S. Ilchenko, H. Mabuchi, et al., Opt. Lett. 23 (1998) 247.
[8] A.A. Savchenkov, V. S Ilchenko, A.B. Matsko, Lute Maleki, Phys. Rev. A 70 (2004) 051804.
[9] J. Yan, J. Tamaki, K. Syoji, T. Kuriyagawa, Int. J. Adv. Manuf. Technol. 24 (2004) 640.
[10] A.E. Fomin, M.L. Gorodetsky, I.S. Grudinin, V.S. Ilchenko, J. Opt. Soc. Am. B 22 (2005) 459.
[11] M.L. Gorodetsky, A.A. Savchenkov, V.S. Ilchenko, Opt. Lett. 21 (1996) 453.
[12] M.L. Gorodetsky, A.D. Pryamikov, V.S. Ilchenko, J. Opt. Soc. Am. B 17 (2000) 1051.
[13] M.L. Gorodetsky, A.E. Fomin, arxiv.org, physics/0509226.
[14] A. Melloni, R. Costa, P. Monguzzi, M. Martinelli, Opt. Lett. 28 (2003) 1567.
[15] V.R. Almeida, C.A. Barrios, R.R. Panepucci, M. Lipson, Nature 431 (2004) 1081.
[16] S.J. Choi, K. Djordjev, et al., IEEE Phot. Technol. Lett. 16 (2004) 828.
[17] A. Savchenkov, I. Grudinin, et al., Opt. (2006) in press.
[18] S.P. Pogossian, L. Vescan, A. Vonsovici, J. Lightwave Technol. 16 (1998) 1851.
[19] W.E. Moerner, M. Orrit, Science 283 (1999) 1670.
[20] W. Kaiser, C.G.B. Garrett, D.L. Wood, Phys. Rev. 123 (1961) 766.
[21] W.E. Moerner, Science 265 (1994) 46.
[22] R.J. Thompson, G. Rempe, H.J. Kimble, Phys. Rev. Lett. 68 (1992) 1132.
[23] M.E. Crenshaw, C.M. Bowden, Opt. Commun. 203 (2002) 115.
[24] V.S. ll'chenko, M.L. Gorodetskii, Laser Physics 2 (1992) 1004.