Ultralow-threshold Raman lasing with CaF_2 resonators

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We demonstrate efficient Raman lasing with CaF_2 whispering-gallery-mode resonators. Continuous-wave emission threshold is shown to be possible below 1 μW with a 5 mm cavity, which is to our knowledge orders of magnitude lower than in any other Raman source. Low-threshold lasing is made possible by the ultrahigh optical quality factor of the cavity, of the order of $Q=5\times10^{10}$. Stokes components of up to the fifth order were observed at a pump power of 160 μW, and up to the eighth order at 1 mW. A lasing threshold of 15 μW was also observed in a 100 μm CaF_2 microcavity. Potential applications are discussed. © 2006 Optical Society of America

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Continuous-wave Raman lasers have drawn considerable attention, as they can provide radiation at virtually any wavelength within a material's spectral transparency window. Generation of stimulated Raman scattering requires high levels of optical power, which may be reduced if a Raman active medium is placed into an optical resonator. In this case, stimulated emission properties are defined by the properties of the resonator, and a Raman laser is formed. Existing configurations include lasers based on variations of Fabry–Perot cavities,[1] Bragg grating resonators,[2] fiber ring cavities,[3] and whispering-gallery-mode (WGM) resonators.[4] The latter exhibit the lowest lasing thresholds, as extremely high optical power density in the cavity is easy to achieve. For example, in a 100 μm fused-silica WGM resonator with $Q=5\times10^8$, an injected power of 1 mW builds up to an intensity of 12 GW/cm^2. Low threshold makes miniaturization of a Raman laser possible. WGM cavities are fiber compatible, compact, and outperform other resonators in terms of lasing efficiency, which make them technologically attractive for building Raman lasers.

The recently demonstrated[5] integrated Raman laser based on a fused-silica microsphere has a lasing threshold of the order of 100 μW, owing mostly to a high intracavity intensity buildup factor. However, the Q factor of fused-silica cavities is subject to degradation due to atmospheric water and is limited to about 8×10^9 for millimeter-sized resonators by material purity achievable with today's technology.

In this Letter we report the demonstration of efficient Raman lasing with ultrahigh-Q crystalline CaF_2 cavities. We also show that achieving a low lasing threshold may be possible by increasing the Q factor as opposed to decreasing the cavity diameter. Indeed, the threshold of Raman lasing in a WGM cavity is given by the following equation[6]:

$$P_{\mathrm{th}}=\frac{\pi^2 n^2}{\xi g_c Q_S Q_P}\frac{V_m}{\lambda_P\lambda_S}. \tag{1}$$

Here, n is the refractive index, and Q_P and Q_S are cavity quality factors for pump and Stokes wavelengths λ_P and λ_S. The cavity Raman gain factor g_c is equal to the bulk Raman gain factor. V_m is the WGM volume, and the factor ξ accounts for noncritical coupling and imperfect overlap of pump and Raman WGMs. If the Q factor is the same for pump and probe wavelengths, the Raman threshold is proportional to the ratio V_m/Q^2. In a realistic cavity the scattering-limited Q factor is proportional to the mode volume $Q\sim V_m$, and the threshold becomes inversely proportional to the latter. From Eq. (1) one can easily see that if a Raman lasing threshold for a 100 μm cavity with $Q=10^8$ is 100 μW, then for a 5 mm cavity with $Q=5\times10^{10}$ the threshold decreases to 0.5 μW. We have recently shown that fabrication of such a cavity, as well as smaller microcavities, is possible with crystalline materials.[7] Fluorite (CaF_2) cavities are immune to water vapor absorption and have stable Q factors. Moreover, ultraefficient single-mode crystalline Raman lasers may now be possible.[8]

We used a recently developed fabrication technique to make two WGM resonators. The technique relies on diamond turning of the crystalline workpiece, followed by a conventional polishing technique. The cavities are made with pure CaF_2 monocrystals and are used as Raman lasing elements. One resonator was made with excimer-grade fluorite with a diameter of 5 mm. The geometry of this multimode resonator is such that its free spectral range (FSR) is 13 GHz, and the cavity supports many nonfundamental WGMs within each FSR. The other cavity was made with a UV-grade fluorite with a diameter of 0.1 mm. The two resonators were cut with their axes parallel to the crystalline 111 orientation. Cavity modes were excited with an angle-polished fiber coupler, which also serves as an output coupler for the pump and Stokes radiation. The output of the coupler was divided between the photodetector and the spectrum analyzer with help of a beam splitter. A Lightwave Electronics Nd:YAG laser with linewidth of less than 5 kHz was used to pump cavity modes. Raman emission was observed with a spectrum analyzer. All experiments were carried out in a clean room, and all measurements were performed for TE

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polarization of the WGMs. To study cw operation of the Raman laser, the Pound-Drever-Hall (PDH) locking technique[9] was implemented to lock the laser frequency to a cavity mode (Fig. 1). A PDH error signal was used to control the laser frequency through an internal piezo element of the laser head. In the process of threshold measurement, the laser was locked to a cavity mode at several pump power values. Pump power refers to the power absorbed in the cavity. The results presented in Fig. 2 correspond to Raman lasing when a WGM with a loaded optical $Q=(2.6\pm0.3)\times10^{10}$ is used, with a coupling efficiency of 40% as measured from the critical coupling point. Clamping of the intracavity power is evident at a point where the second Stokes component appears. This behavior is natural for WGM-based Raman lasers, as was shown elsewhere.[5] Given the threshold of 3 μW at this Q factor and Eq. (1) we see that if a different mode with a Q factor of 50×10^9 and the same effective volume is used, a threshold well below 1 μW should be expected. It was shown[5] that the optimal coupling for Raman lasing is about 89% of critical, which also implies some room for further optimization. We believe this makes the crystalline WGM resonator the most efficient Raman source available today, as the threshold of other sources is at least 20 times higher. The efficiency of our laser was made small by operating in an undercoupled regime with the purpose of measuring the lasing threshold. The maximum observed unidirectional efficiency for the first Stokes component was $P_{\text{Stokes}}/P_{\text{pump}}=0.24$ in cascaded mode with seven Stokes components present.

The vibrational spectrum of CaF_2 has nine phonon branches—six optical and three acoustic. Only one vibration with $\nu=322\ \text{cm}^{-1}$ is Raman active and corresponds to vibrations of positive fluorine ions. The Raman wavenumber and gain linewidth depend on temperature.[10,11] The wavelengths of the Stokes components may be computed and are found to be in direct agreement with the experimental results presented in Fig. 3. The inset shows the first Stokes spectrum, which is formed by many WGMs lasing within each FSR of the cavity. The spectrum width gives a rough estimate for the Raman gain linewidth $\delta\nu\simeq1.7\ \text{cm}^{-1}$.

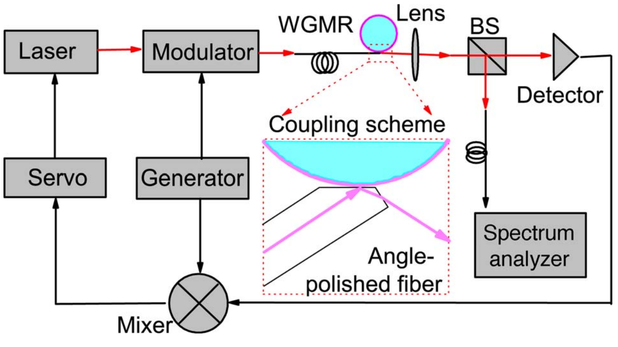

Fig. 1. (Color online) Setup diagram with (inset) coupling schematics. A PDH locking technique is used to stabilize the power in a cavity. WGMR, whispering-gallery-mode resonator; BS, beam splitter.

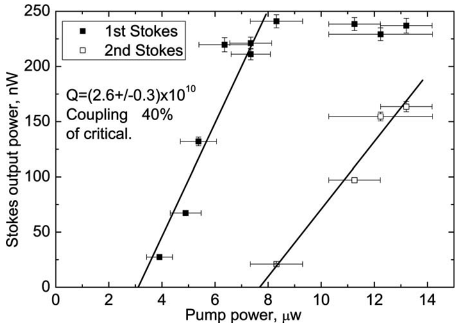

Fig. 2. Output optical power as a function of the pump power for nonoptimized operation of the Raman laser based on a 5 mm cavity.

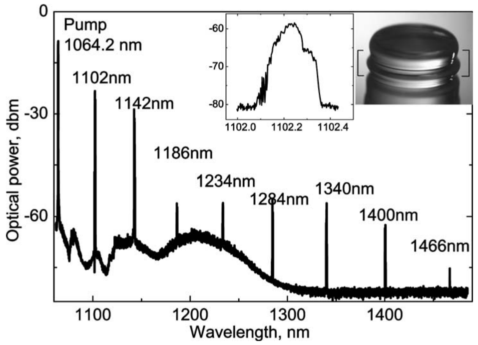

Fig. 3. Cascaded operation of the Raman laser. Insets, photograph of a cavity and detailed spectrum of the first Stokes component. Power is as seen on the spectrum analyzer; the actual pump power is 1 mW.

Cascaded operation of the laser with Stokes components of up to the eighth order was observed in an unlocked regime, when the pump laser frequency is quickly scanned around the WGM. A high-mode pump power of about 1 mW produced a nonlinear behavior of the mode so pronounced that the PDH locking was impossible. As a consequence, the power levels of Stokes components in Figs. 3 and 4 were fluctuating over the time of the measurement. These components indicate only the presence of Raman emission at the given wavelengths. Cascaded Raman lasing has been previously observed in microdroplets and, for example, in liquid hydrogen droplets[12] at rather high-input power levels. Efficient Raman lasing with five Stokes orders was also observed in fused-silica microcavities.[4] We present what we believe to be the first observation of cascaded operation of CaF_2 Raman laser and the first observation of eight Stokes components in a fiber-compatible all-solid-state crystalline laser at a pump power below 1 mW.

We have also observed Raman emission from a smaller cavity made with a different specimen of CaF_2. It is interesting to compare the operation of our laser with the one discussed in Ref. 4. The microsphere with the lowest threshold had a diameter of about 55 μm, a quality factor $Q=10^8$, and a first Stokes threshold of about 60 μW. In the fluorite cavity presented here (Fig. 5), the modes occupy a larger space as the resonator diameter is 100 μm. The value of the quality factor $Q=10^8$ is obtained with direct linewidth measurements. The Raman lasing threshold in the fluorite cavity is estimated to be below 15 μW, which may suggest a stronger Raman gain in CaF_2. This argument is supported by a study of scattering in fluorite,[13] where the gain for CaF_2 was found to be higher as compared with fused silica, while having a much narrower spectral width. At a pump power of about 100 μW cascaded operation of the Raman microlaser is observed. No anti-Stokes emission was found for our cavities.

In summary, we have demonstrated that WGM resonators made with crystalline CaF_2 may be used as efficient fiber-compatible Raman lasing sources with ultralow thresholds. The low Raman lasing threshold and efficient cascaded operation are made possible by the ultrahigh optical quality factors of the WGMs of crystalline cavities. For a 5 mm cavity diameter, the threshold of the first Stokes was demonstrated to be as low as 3 μW. The prospect for the submicrowatt threshold was also discussed. Cascaded operation with eight Stokes components is observed for what we believe to be the first time with only 1 mW of pump power. The wide variety of optical crystals in combination with crystalline WGM technology make compact, fiber-compatible tunable Raman lasers possible. Fiber compatibility is provided by the coupler we use, which may be viewed as a fiber–prism hybrid. Although coupling was not optimized in this work, we routinely observe more than 80% coupling efficiency with angle-polished fiber couplers for any resonator diameters. Stokes emission could also be improved if a second angle-optimized coupler is used. By a proper choice of the nonlinear crystal, one may achieve a generation of many different Stokes components with low-threshold power and high efficiency.

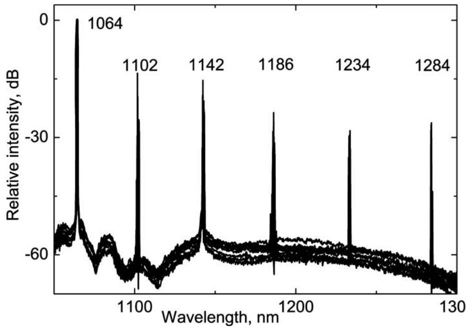

Fig. 4. Cascaded operation of the Raman laser. The pump power is 160 μW. The spectrum is combined from nine consecutive measurements obtained in an unlocked regime.

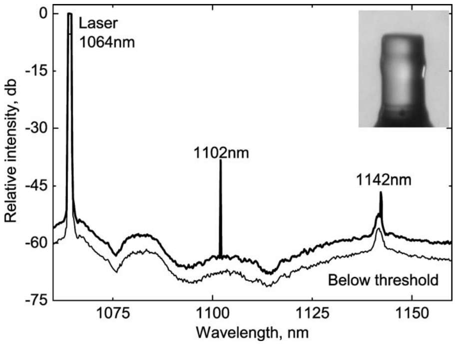

Fig. 5. Cascaded Raman lasing in a 0.1 mm fluorite microcavity. The pump power is about 100 μW.

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