Low Threshold Optical Oscillations in a Whispering Gallery Mode CaF_2 Resonator

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We have observed low-threshold optical hyperparametric oscillations in a high-Q fluorite whispering gallery mode resonator. The oscillations result from the resonantly enhanced four-wave mixing occurring due to Kerr nonlinearity of the material. We demonstrate that, because of the narrow bandwidth of the resonator modes as well as the high efficiency of the resonant frequency conversion, the oscillations produce stable narrow-band beat-note of the pump, signal, and idler waves. A theoretical model for this process is described.

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Realization of efficient nonlinear optical interactions at low light levels has been one of the main goals of nonlinear optics since its inception. Optical resonators contribute significantly to achieving this goal, because confining light in a small volume for a long period of time leads to increased nonlinear optical interactions. Optical whispering gallery mode (WGM) resonators are particularly well suited for the purpose. Features of high quality factors (Q) and small mode volumes have already led to the observation of low-threshold lasing as well as efficient nonlinear wave mixing in WGM resonators made of amorphous materials [1,2].

This Letter is devoted to the study of optical hyperparametric processes in a WGM resonator made of CaF_2—a low loss crystalline material possessing cubic nonlinearity. Optical hyperparametric oscillations, dubbed as modulation instability in fiber optics, usually are hindered by small nonlinearity of the materials, so high-power light pulses are required for their observation [3]. Though the nonlinearity of CaF_2 is even smaller than that of fused silica, we were able to observe with low-power continuous wave pump light a strong nonlinear interaction among resonator modes resulting from the high Q ($Q > 5 \times 10^9$) of the resonator. New fields are generated due to this interaction.

We show that the frequency of the microwave signal produced by mixing the pump and the generated sidebands on a fast photodiode is very stable and does not experience a frequency shift that could occur due to the self- and cross-phase modulation effects. Conversely in, e.g., coherent atomic media, the oscillations frequency shifts to compensate for the frequency mismatch due to the cross-phase modulation effect (ac Stark shift) [4,5]. In our system the oscillation frequency is given by the mode structure and, therefore, can be tuned by changing the resonator dimensions. Finally, in contrast with resonators fabricated with amorphous materials and liquids, high-Q crystalline resonators allow for a better discrimination of the third-order nonlinear processes and the observation of pure hyperparametric oscillation signals. As a result, the hyperoscillator is promising for applications as an all-optical secondary frequency reference.

The hyperparametric oscillations could be masked with stimulated Raman scattering (SRS) and other nonlinear effects. For example, an observation of secondary lines in the vicinity of the optical pumping line in the SRS experiments with WGM silica microresonators was interpreted as four-wave mixing between the pump and two Raman waves generated in the resonator, rather than as the four-photon parametric process based on electronic Kerr nonlinearity of the medium [6]. An interplay among various stimulated nonlinear processes has also been observed and studied in droplet spherical microcavities [7].

The polarization selection rules together with WGM's geometrical selection rules allow for the observation of nonlinear processes occurring solely due to the electronic nonlinearity of the crystals in crystalline WGM resonators. Let us consider a fluorite WGM resonator possessing cylindrical symmetry with [1,0,0] symmetry axis. The linear index of refraction in a cubic crystal is uniform and isotropic [8], therefore the usual description of the modes [1] is valid for the resonator. The TE and TM families of WGMs have polarization directions parallel and orthogonal to the symmetry axis, respectively. If an optical pumping light is sent into a TE mode, the Raman signal cannot be generated in the same mode family because in a cubic crystal such as CaF_2 there is only one, triply degenerate, Raman-active vibration with symmetry F_{2g} [9]. Finally, in the ultrahigh Q crystalline resonators, due to the material as well as geometrical dispersion, the value of the free spectral range (FSR) at Raman detuning differs from the FSR at the carrier frequency by an amount exceeding the mode spectral width. Hence, frequency mixing between the Raman signal and the carrier is strongly suppressed. Any field generation in the TE mode family is due to the electronic nonlinearity only, and Raman scattering occurs in the TM modes.

The scheme of our experimental setup is shown in Fig. 1. Light from a 1.32 μm YAG laser is sent into a CaF_2 WGM resonator with a glass coupling prism. The laser linewidth is less than 5 kHz. The maximum coupling efficiency is better than 50%. A typical CaF_2 resonator has a toroidal shape with a diameter of several millimeters and thickness in the range of several hundred microns. The resonators Q factors are on the order of 10^9–10^{10}.

The output light of the resonator is collected into a single-mode fiber after the coupling prism, and is split into two equal parts with a 50/50 fiber splitter. One output of the splitter is sent to a slow photodiode $D1$ that produces a dc signal used for locking the laser to a particular resonator's mode. The other output is mixed with a delayed laser light that has not interacted with the resonator, and the mixed signal is directed to fast photodiode $D2$. With this configuration the disc resonator is placed into an arm of a tunable Mach-Zehnder interferometer. If the delay between the interferometer arms is correctly chosen we observe a narrow-band microwave signal emitted by the photodiode.

The locking loop enables us to inject a desired amount of optical power into the resonator, which would be difficult otherwise because the resonator spectrum can drift due to thermal and Kerr effects. The larger the laser power, the further do the modes shift, reducing total power accumulation in the resonator. For instance, thermal dependence of the index of refraction for CaF_2 is $\beta = n_0^{-1}\partial n/\partial T \simeq -10^{-5}$/K. This means that the frequency of a WGM mode ω_m increases by $10^{-5}\omega_m$ if the temperature T increases by 1 ° Kelvin (follows from $\omega_m \approx mc/Rn_0(1+\beta)$, where c is the speed of light in the vacuum, $m \gg 1$ is the mode number, R is the radius of the resonator, and n_0 is the index of refraction). Such a shift is 4 orders of magnitude larger than the width of the resonance if $Q = 10^9$. The feedback loop compensates for this shift by adjusting the laser frequency to keep up with the mode. The stationary mode frequency is determined by the amount of the optical power absorbed in the resonator as well as by the heat exchange of the cavity with the external environment.

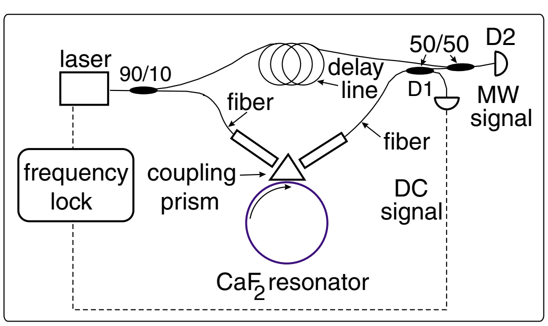

FIG. 1 (color online). Experimental setup.

We found that if the light from the resonator is directly sent to a fast photodiode it would not generate any detectable microwave signal. However, if the resonator is placed into an arm of the Mach-Zehnder interferometer and the delay in the second arm of the interferometer is correctly chosen, the modulation appears. This is a distinct property of phase modulated light. The observed phase modulation implies that high- and low-frequency sidebands generated in the parametric oscillation process have a π radian phase relationship, while the hyperparametric oscillations observed in optical fibers generally result in a $\pi/2$ phase between the sidebands [10].

The typical microwave spectrum is depicted in Fig. 2. The generated microwave signal with the frequency ~8 GHz corresponding to the FSR of the resonator has a narrow ($\leq$ 40 kHz) linewidth. We found that the signal frequency is stable with temperature, pump power, and coupling changes, because the FSR frequency of the resonator does not significantly change with any of those parameters.

We measured the value of the microwave power as a function of the optical pump power (Fig. 3) to find the efficiency of the parametric process. The modulation appears only after exceeding a distinct power threshold (~1 mW) for the optical pump. We estimated the modulation efficiency to be ~7% between a sideband power and the pump power achieved at 4 mW input optical power. Increasing the pumping power results in a gradual decrease of the sideband power because of the generation of higher harmonics, see in Fig. 3.

We also searched for the SRS process in the resonator. To collect all output light from the TE as well as the TM modes we used a multimode optical fiber instead of the single-mode fiber at the output of the prism coupler. The fiber was connected to an optical spectrum analyzer. The SRS signal was expected in the vicinity of the CaF_2

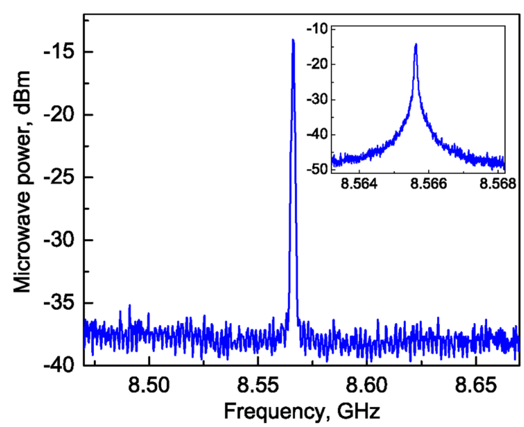

FIG. 2 (color online). 22 dB amplified microwave signal generated at the broadband optical detector by the light interacted with a CaF_2 resonator. The microwave frequency corresponds to FSR of the resonator. The half width at half maximum of the signal shown is less than 40 kHz.

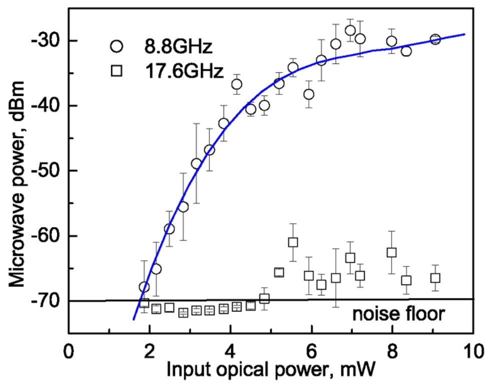

FIG. 3 (color online). Plot of the microwave power at the output of the optical detector versus 1.32 μm pumping light power. The microwave frequency corresponds to the $\delta_{\rm FSR}$ of the disc. The solid line is a guide for the eye. Circles and squares are for the first and second mw harmonics of the signal.

Raman-active phonon mode (322 cm^{-1} [11]). Within the range of accuracy of our measurement setup and with optical pumping as high as 10 mW at the resonator entrance, we were unable to observe any SRS signals, in contrast with the previous studies of nonlinear phenomena in amorphous WGM resonators [6,7]. We conclude that the modulation effect is due to the hyperparametric oscillations, not a four-wave mixing between the optical pumping and light generated due to the Raman scattering.

To explain the results of our experiment we consider three cavity modes: one nearly resonant with the pump laser and the other two nearly resonant with the generated optical sidebands. We begin with the following equations for the slow amplitudes of the intracavity fields

$$\dot{A} = -\Gamma_0 A + ig[|A|^2 + 2|B_+|^2 + 2|B_-|^2]A + 2igA^* B_+ B_- + F_0, \tag{1}$$

$$\dot{B}_+ = -\Gamma_+ B_+ + ig[2|A|^2 + |B_+|^2 + 2|B_-|^2]B_+ + igB_-^*|A|^2, \tag{2}$$

$$\dot{B}_- = -\Gamma_- B_- + ig[2|A|^2 + 2|B_+|^2 + |B_-|^2]B_- + igB_+^*|A|^2, \tag{3}$$

where $\Gamma_0 = i(\omega_0 - \omega) + \gamma_0$ and $\Gamma_\pm = i(\omega_\pm - \tilde{\omega}_\pm) + \gamma_\pm$, γ_0, γ_+, and γ_- as well as ω_0, ω_+, and ω_- are the decay rates and eigenfrequencies of the optical cavity modes respectively; ω is the carrier frequency of the external pump (A), $\tilde{\omega}_+$ and $\tilde{\omega}_-$ are the carrier frequencies of generated light (B_+ and B_-, respectively). These frequencies are determined by the oscillation process and cannot be controlled from the outside. However, there is a relation between them (energy conservation law): $2\omega = \tilde{\omega}_+ + \tilde{\omega}_-$. Dimensionless slowly varying amplitudes A, B_+, and B_- are normalized such that $|A|^2$, $|B_+|^2$, and $|B_-|^2$ describe photon number in the corresponding modes. The coupling constant can be found from expression $g = \hbar\omega_0^2 n_2 c/\mathcal{V} n_0^2$, where n_2 is an optical constant that characterizes the strength of the optical nonlinearity, n_0 is the linear refractive index of the material, $\mathcal{V}$ is the mode volume, and c is the speed of light in the vacuum. Deriving this coupling constant we assume that the modes are nearly overlapped geometrically, which is true if the frequency difference between them is small. The force F_0 stands for the external pumping of the system $F_0 = (2\gamma_0 P_0/\hbar\omega_0)^{1/2}$, where P_0 is the pump power of the mode applied from the outside.

For the sake of simplicity we assume that the modes are identical, i.e., $\gamma_+ = \gamma_- = \gamma_0$, which is justified by observation with actual resonators. Then, solving the set (1)–(3) in steady state we find the oscillation frequency for generated fields

$$\omega - \tilde{\omega}_- = \tilde{\omega}_+ - \omega = \frac{1}{2}(\omega_+ - \omega_-), \tag{4}$$

i.e., the beat-note frequency depends solely on the frequency difference between the resonator modes and does not depend on the light power or the laser detuning from the pumping mode. As a consequence, the electronic frequency lock circuit changes the carrier frequency of the pump laser but does not change the frequency of the beat note of the pumping laser and the generated sidebands.

The threshold optical power can be found from the steady state solution of the set (1)–(3)

$$P_{\rm th} \simeq 1.54\frac{\pi}{2}\frac{n_0^2\mathcal{V}}{n_2\lambda Q^2}, \tag{5}$$

where the numerical factor 1.54 comes from the influence of the self-phase modulation effects on the oscillation threshold. The theoretical value for threshold in our experiment is $P_{\rm th} \approx 0.3$ mW, where $n_0 = 1.44$ is the refractive index of the material, $n_2 = 3.2 \times 10^{-16}$ cm^2/W is the nonlinearity coefficient for calcium fluoride, $\mathcal{V} \approx 10^{-4}$ cm^3 is the mode volume, $Q = 6 \times 10^9$, and $\lambda = 1.32$ μm.

It is easy to see from Eq. (5) that the efficiency of the parametric process increases with a decrease of the mode volume. We used a relatively large WGM resonator because of the fabrication convenience. Reducing the size of the resonator could result in a dramatic reduction of the threshold for the oscillation. Since the mode volume may be roughly estimated as $\mathcal{V} \approx 2\pi\lambda R^2$, it is clear that reducing the radius R by an order of magnitude would result in 2 orders of magnitude reduction in the threshold of the parametric process. This could place WGM resonators in the same class as the oscillators based on atomic coherence [4]. However, unlike the frequency difference between sidebands in the atomic oscillator, the frequency of the WGM oscillator could be free from power (ac Stark) shifts.

Solving the set of Langevin equations describing quantum behavior of the system we found that the phase diffusion of the beat-note is small, similar to the low phase diffusion of the hyperparametric process in atomic coherent media [5]. Close to the oscillation threshold the phase diffusion coefficient is

$$D_{\mathrm{beat}} \simeq \frac{\gamma_0^2}{4} \frac{\hbar\omega_0}{P_{B\,\mathrm{out}}}, \tag{6}$$

where $P_{B\,\mathrm{out}}$ is the output power in a sideband. Allan deviation is $\sigma_{\mathrm{beat}}/\omega_{\mathrm{beat}} = (2D_{\mathrm{beat}}/t\omega_{\mathrm{beat}}^2)^{1/2}$. We could estimate $\sigma_{\mathrm{beat}}/\omega_{\mathrm{beat}} \simeq 10^{-13}/\sqrt{t}$ for $\gamma_0 = 3 \times 10^5$ rad/s, $P_{B\,\mathrm{out}} = 1$ mW, $\omega_0 = 1.4 \times 10^{15}$ rad/s, and $\omega_{\mathrm{beat}} = 5 \times 10^{10}$ rad/s. Follow up studies of the stability of the oscillations in the general case will be published elsewhere.

We considered only three interacting modes in the model, however the experiments show that a larger number of modes could participate in the process. The number of participating modes is determined by the variation of the mode spacing in the resonator. Generally, modes of a resonator are not equidistant because of the second order dispersion of the material and the geometrical dispersion. We introduce $D = (2\omega_0 - \omega_+ - \omega_-)/\gamma_0$ to take the second order dispersion of the resonator into account. If $|D| \geq 1$ the modes are not equidistant and, therefore, multiple harmonic generation is impossible.

Geometrical dispersion for the main mode sequence of a WGM resonator is $D \simeq 0.41c/(\gamma_0 R n_0 m^{5/3})$, for a resonator with radius R; ω_+, ω_0, and ω_- are assumed to be $m+1$, m, and $m-1$ modes of the resonator ($\omega_m R n_{\omega_m} = mc$, $m \gg 1$). For $R = 0.4$ cm, $\gamma_0 = 2 \times 10^5$ rad/s, $m = 3 \times 10^4$ we obtain $D = 7 \times 10^{-4}$, therefore the geometrical dispersion is relatively small in our case. However, the dispersion of the material is large enough. Using the Sellmeier dispersion equation [8] we find $D \simeq 0.1$ at the pump laser wavelength. This implies that approximately three sideband pairs can be generated in the system (we see only two in the experiment).

Finally, the absence of the Raman signal in our experiments shows that effective Raman nonlinearity of the medium is lower than the value measured earlier [9]. Theoretical estimates based on numbers from [9] predict nearly equal pump power threshold values for both the Raman and the hyperparametric processes. Using the expression derived for SRS threshold (see in [6]) $P_R \simeq \pi^2 n_0^2 \mathcal{V}/G\lambda^2 Q^2$, where $G \simeq 2 \times 10^{-11}$ cm/W is the Raman gain coefficient for CaF_2, we estimate $P_{\mathrm{th}}/P_R \approx 1$ for any resonator made of CaF_2. However, as mentioned above, we did not observe any SRS signal in the experiment.

In conclusion, we have observed and analyzed optical parametric oscillations in a high-Q crystalline resonator. Because of the long interaction times of the pumping light with the material, even the small cubic nonlinearity of CaF_2 results in an efficient generation of narrow-band optical sidebands. This process can be used for the demonstration of a new kind of an all-optical frequency reference. Moreover, the oscillations are promising as a source of squeezed light because the sideband photon pairs generated in the hyperparametric processes are generally quantum correlated.

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Note added.—A study of Kerr-nonlinearity based optical parametric oscillations in a fused silica toroid microcavity was recently reported by Kippenberg *et al.* in [12].

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