Nonlinear Optics and Crystalline Whispering Gallery Mode Cavities

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We demonstrate parametric frequency doubling in a whispering gallery cavity made of periodically poled lithium niobate. This demonstration is an example of utility of such crystalline optical whispering gallery resonators with very high Q factors, which we have fabricated.

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Dielectric cavities with whispering gallery (WG) modes are unique because of their ability to store light in microscopic spatial volumes for long periods of time, resulting in strong enhancement of nonlinear interactions of various kinds [1–7]. High Q WG modes were first observed in spherical droplets of liquids [1,3], as well as in solidified droplets of fused amorphous materials [8–11]. Although these materials possess very small optical attenuation, the highest quality factor of WG modes has remained limited by Rayleigh scattering of residual surface roughness [12]. This is generally the case, even though a cavity formed by surface tension forces has nearly a defect-free surface characterized by molecular-scale inhomogeneities. Any residual roughness would result in serious limitation on the achievable cavity Q factor. Hence, it is commonly believed that high Q optical WG cavities cannot be fabricated with transparent crystals. Melting is obviously not suitable for materials with crystalline structure, because it destroys the initial crystal purity and stoichiometry. Moreover, during solidification, the original spherical droplet of the melt turns into a rough body with multiple facets and crystal growth steps.

In this work we demonstrate that it is possible to obtain crystalline WG optical resonators with very high Q factors ($Q > 10^8$), similar to that of surface-tension-formed resonators, by adopting simple polishing techniques. With this approach, the original crystal structure and composition are preserved, and the unique linear and nonlinear crystal properties are enhanced with the small volume of the high Q cavity. Total internal reflection at the walls of the WG cavities provides the effect of an ultrabroad band mirror, allowing very high Q factors across the whole material transparency range. This property makes WG crystal cavities a unique tool for optical materials studies. With our fabrication process, we have achieved a Q factor limited in value only by the absorption of the material. As an example of the potential of WG crystal cavities for nonlinear optics applications, we demonstrated parametric frequency doubling in a WG cavity made of periodically poled lithium niobate.

Crystals generally have high purity, a high index of refraction, and a high stability of structure, and are thus ideal for WG cavities. Ultimate Q factors of WG cavities made of fused silica degrade in the atmosphere due to diffusion of atmospheric water into the material [9]. Water molecules, however, cannot diffuse into some crystals. Typically, crystals also have very low intrinsic absorption of light. For example, absorption of sapphire determined by light scattering due to imperfection of the crystalline structure is less than $\alpha = 1.3 \times 10^{-5}$ cm^{-1} at $\lambda = 1$ μm [13] which corresponds to $Q \simeq 8 \times 10^9$. Light absorption for the crystalline quartz may also be estimated using absorption in fused silica, which is extensively studied for fiber optic applications, with $\alpha \leq 5 \times 10^{-6}$ cm^{-1} at $\lambda = 1.55$ μm [14]; this corresponds to $Q \geq 1.2 \times 10^{10}$ for quartz.

A particular advantage of crystalline cavities is that they enable a variety of studies with high nonlinearity optical materials. For example, resonant interaction of light and microwave was recently achieved using a WG optical cavity made of $LiNbO_3$. A new kind of electro-optic modulator and photonic receiver based on this interaction was also suggested and realized [15,16]. Coupling several crystalline cavities to create microstructured resonator systems could further enhance nonlinear properties of the crystals and, hence, has a lot of potential photonics applications [17–19].

The efficiency of nonlinear optical processes increases with the increasing Q factor of the modes of WG cavities. The maximum quality factor of WG modes in lithium niobate cavity reported in [16] was less than 5×10^6 at $\lambda = 1.55$ μm, which approximately corresponded to values expected from the intrinsic absorption of congruent lithium niobate cited by crystal producers ($\alpha \leq 5 \times 10^{-3}$ cm^{-1}). On the other hand, $Q \approx 2 \times 10^8$ at $\lambda = 2.014$ μm ($\alpha \leq 5 \times 10^{-4}$ cm^{-1}) was reported earlier for a multiple total-internal-reflection resonator, analogous to a WG cavity, used in optical parametric oscillators pumped at 1064 nm [20]. This suggests that the Q factors achieved with WG mode cavitities were limited by the fabrication process, and that the above values for the absorption of congruent lithium niobate may be inaccurate.

We succeeded in fabricating toroidal lithium niobate WG mode cavities with quality factors as high as $Q = 2 \times 10^8$ at $\lambda = 1.31$ μm, using diamond polishing of the rim of flat disk preforms. The disks typically have a radius of 0.75–7.5 mm and thickness of 0.05–1 mm.

Our measurement lowers the experimentally achieved optical absorption floor of congruent $LiNbO_3$ up to $\alpha \leq 4 \times 10^{-4}$ cm^{-1} at that wavelength. We have also obtained $Q = 5 \times 10^7$ at $\lambda = 0.78$ μm.

Preliminary experiments show that our polishing technique is applicable not only to the lithium niobate, but also to a variety of other crystalline materials, ranging from crystalline quartz to sapphire. For instance, we reached $Q = 6.2 \times 10^8$ at $\lambda = 1.31$ μm and $Q = 8 \times 10^7$ at $\lambda = 0.78$ μm in a sapphire resonator. This makes the WG cavity a more general tool for measurement of small optical losses and other properties of crystals, as well as a versatile building block for novel experiments in quantum and nonlinear optics.

We achieved efficient frequency doubling at $\lambda = 1.55$ μm and $\lambda = 1.319$ μm using the same WG cavity made of periodically poled $LiNbO_3$ (PPLN) [21]. The cavity is doubly resonant, both at fundamental and second harmonic frequencies. It is interesting to note that Q factors are the same for both the PPLN cavities and our original congruent lithium niobate cavities—proving that the periodical poling does not change linear absorption of the material. The follow-up studies of the parametric processes in WG PPLN cavities are important because it has been predicted that an optical parametric oscillator (OPO) based on a WG PPLN cavity might have a power threshold below a microwatt [22]—orders of magnitude less than that of the state-of-the-art OPOs, typically at the 0.5 mW level [23].

Frequency conversion may be realized when energy and momentum conservation (phase matching condition) for photons are fulfilled during the nonlinear interaction. For example, the energy conservation law gives a trivial condition for frequency doubling $\lambda_p = 2\lambda_s$, where λ_p and λ_s are the pump and signal wavelengths in vacuum. On the other hand, momentum conservation is not generally satisfied in crystals: $2k_p - k_s \neq 0$, where k_p and k_s are wave vectors of the pump and signal waves. This problem may be handled using a quasiphase matching technique [21], which is based on producing a modulation of the nonlinear susceptibility of the material with a period Λ such that

$$k_s - 2k_p = \frac{2\pi}{\Lambda} \rightarrow \Lambda = \frac{\lambda_s}{n_s - n_p}, \tag{1}$$

where n_s and n_p are indices of refraction for the signal and pump light. Spatial modulation of nonlinear susceptibility may be obtained by local flipping of the sign of the spontaneous polarization, i.e., by flipping the spins in ferroelectric domains. From the point of view of optical nonlinearity, oppositely polarized domains exhibit opposite signs of second-order susceptibility [21].

To achieve quasiphase matching for parametric frequency doubling in a WG cavity, one should take into account not only the frequency dependent dispersion of the host material of the dielectric cavity, but also the dispersion introduced by the internal geometrical mode structure. Modes inside a WG cavity are localized close to the cavity rim. The shorter the wavelength, the closer is the mode to the cavity surface, and the longer is the mode path. This geometrical property of WG modes significantly changes the phase matching condition compared with bulk material.

The frequency of the main sequence of high order TE WG modes may be estimated from

$$\frac{2\pi R}{\lambda} n(\lambda) + \sqrt{\frac{n^2(\lambda)}{n^2(\lambda) - 1}} \simeq \nu + \alpha_q \left(\frac{\nu}{2}\right)^{1/3} + \frac{3\alpha_q^2}{20}\left(\frac{2}{\nu}\right)^{1/3}, \tag{2}$$

where λ is the wavelength in vacuum, ν is the mode order, $n(\lambda)$ is the wavelength dependent index of refraction, R is the radius of the cavity, and α_q is the qth root of the Airy function, $Ai(-z)$, which is equal to 2.338, 4.088, and 5.521 for $q = 1, 2, 3$, respectively [24]. The phase matching condition (1) should be rewritten here as $\nu_s - 2\nu_p = \nu_\Lambda$, where ν_s, ν_p, and ν_Λ are numbers of signal, pump, and the nonlinearity modulation (i.e., number of poling periods in the cavity) modes.

In our experiments, we used a WG cavity fabricated from a commercial flat Z-cut $LiNbO_3$ substrate, with TE modes corresponding to the extraordinary waves in the material. The cavity has a radius of $R = 1.5$ mm and thickness of $d = 0.5$ mm. The curvature of the rim is approximately 1.2 mm. The lithium niobate substrate is periodically poled with $\Lambda = 14$ μm period. The poling is made in stripes, as schematically shown in Fig. 1(c).

Because WG modes are localized close to the surface of the cavity, they encounter many periods of nonlinearity modulation, not just a single period. This provides us with the opportunity to achieve frequency doubling at a wide range of frequencies including those that do not correspond to the original planar poling period. We found that, for the cavity that we are using in the experiment, the amplitudes of the corresponding harmonics of

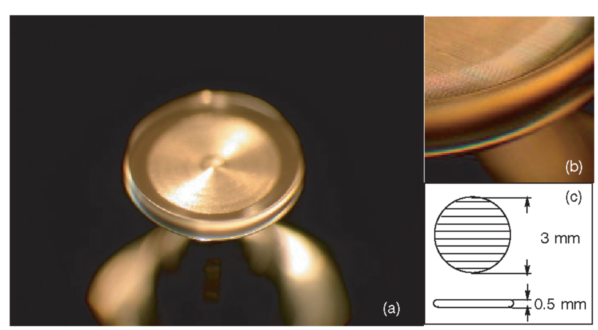

FIG. 1 (color online). (a) Toroidal whispering gallery mode cavity made of periodically poled lithium niobate. (b) Enlarged fragment of the cavity rim. (c) Schematic of the cavity. Poling has planar structure and 14 μm period.

the nonlinearity of the poled crystal are $\chi^{(2)}_{1.55} \simeq 1.6 \times 10^{-3}\chi^{(2)}\cos(\nu_{1.55}\phi)$ (phase-matched harmonic of the nonlinearity for 1550 nm pump) and $\chi^{(2)}_{1.319} \simeq 8 \times 10^{-5}\chi^{(2)}\cos(\nu_{1.319}\phi)$ (phase-matched harmonic of the nonlinearity for 1319 nm pump), where $\chi^{(2)}$ is the nonlinearity of the material. We observed doubling for both of these frequencies. The efficiency of the frequency doubling at $\lambda = 1.55$ μm is shown in Fig. 2.

The absorption spectrum of the cavity measured by frequency scanning of the pump laser at 1.55 μm and the emission spectrum for the second harmonic at 775 nm, taken simultaneously, are shown in Fig. 3. Light from a pump laser is sent into the cavity via a coupling diamond prism. The prism output is collected to a multimode fiber. Using spatial selection and a photodiode detecting the second harmonic only, we are able to filter out the residual pump light going out of the prism. It is not so difficult to find a value of the pump frequency for which the second harmonic is also resonant in the cavity we used. It is interesting to note that phase matching conditions are not periodic with the cavity free spectral range (FSR). The cavity FSR (13.6 GHz) is easily identifiable through the periodic pattern of the absorption peaks in Fig. 3. Only one mode for the second harmonic is efficiently coupled to the pump, within a more than 80 GHz scan of the pump laser—roughly 6 times the free spectral range. The bandwidth of the second harmonic nonlinear resonance, in units of pump laser frequency tuning, is approximately 5 MHz. The maximum signal power on the prism output observed in the experiment was approximately 12.3 mW. The pump power at this point was 25 mW at the input port of the cavity, i.e., the maximum conversion efficiency was approximately 0.5. For the frequency doubling at $\lambda = 1.319$ μm, the maximum efficiency was 2×10^{-2} for the pump power available (30 mW). This relatively small efficiency at 1.319 μm may be explained by the small effective value of nonlinearity or, in other words, the poor efficacy of the chosen grating poling period.

The conversion efficiency of the frequency doubling can be found theoretically and compared with the experiment (see, for example, [22,25]). The interaction Hamiltonian in slowly varying amplitude and phase approximation is

$$H = \hbar g[(a_p^\dagger)^2 a_s + a_s^\dagger a_p^2], \tag{3}$$

where the coupling constant is

$$g = 2\pi\omega_p \frac{\chi^{(2)}_{1.55}}{\epsilon_p}\frac{V_{spp}}{V_p}\sqrt{\frac{2\pi\hbar\omega_s}{\epsilon_s V_s}}, \tag{4}$$

$V_{spp} = \int_V \Psi_s \Psi_p^2 dV < (V_p, V_s)$ is the mode overlap integral, and a_p ($a_p^\dagger$) and a_s ($a_s^\dagger$) are mode annihilation (creation) operators for the signal and pump electromagnetic field in the cavity modes.

Using this Hamiltonian, we derive equations of motion, find the photon number for the second harmonic generated in the system, and derive the output power of the signal with respect to the pump power

$$\frac{W_{s\ \mathrm{out}}}{W_{p\ \mathrm{in}}} = \frac{6}{S}\{[1 + S + \sqrt{S(1+S)}]^{1/3} + [1 + S - \sqrt{S(1+S)}]^{1/3} - 2\}^2, \tag{5}$$

where $S = 54W_{p\ \mathrm{in}}/W_0$ is the saturation parameter, and

$$W_0 = \frac{n_p^4 n_s^2}{64\pi^3[\chi^{(2)}_{1.55}]^2}\left(\frac{V_p}{V_{pps}}\right)^2\frac{\omega_p V_s}{Q_p^2 Q_s} \tag{6}$$

is the saturation power.

For our experimental parameters [$V_{pps}/V_p = 0.3$, $V_s \approx 2\pi R \times 2R(2\pi/\nu_s)^{1/2} \times (R/\nu_s^{2/3}) = 10^{-6}$ cm^3, $n_p = 2.138$, $n_s = 2.179$, $\chi^{(2)}_{1.55} = 7 \times 10^{-10}$ cgs, $\omega_p = 2 \times 10^{15}$ s^{-1}, loaded Q factors: $Q_s \simeq 8 \times 10^6$ and $Q_p \simeq 1.2 \times 10^7$],

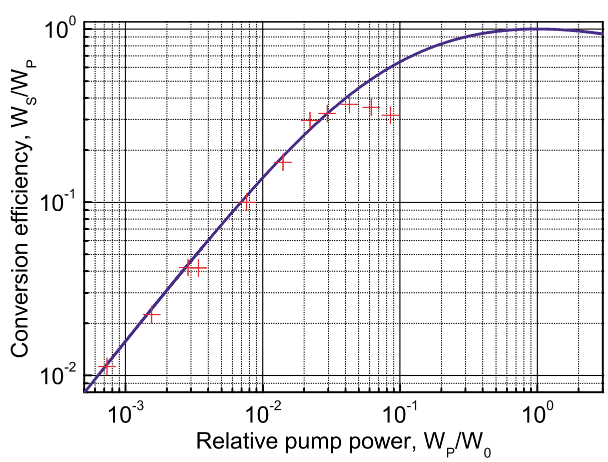

FIG. 2 (color online). Conversion efficiency as function of normalized input pump power. Experimentally measured saturation parameter W_0 is 0.3 W.

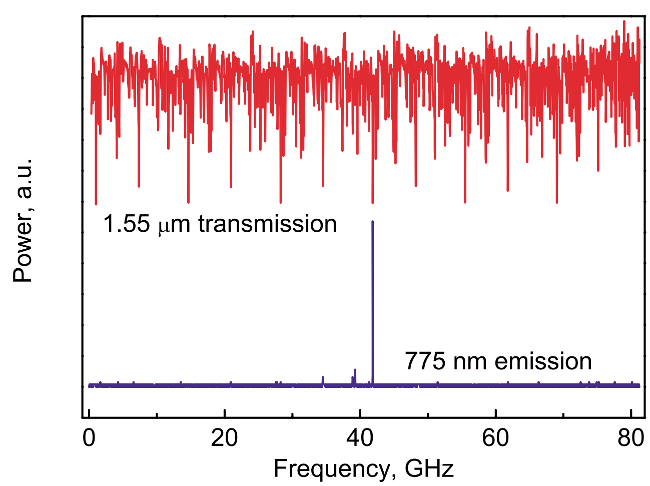

FIG. 3 (color online). Pump transmission and signal emission spectra of the cavity.

the calculated value of the saturation power is 190 mW. The 300 mW value for the saturation power was derived from our experimental data. It can easily be seen that increasing the Q factors of the modes and producing exact periodical poling for the appropriate wavelengths will result in orders of magnitude reduction of the required pump power, while keeping the high conversion efficiency intact.

The conversion efficiency in our experiment saturates faster than the theoretical prediction. We believe this effect is due to temperature fluctuations. We were unable to keep the temperature stable for the applied high laser power (the pump intensity in the cavity could be as high as $10\ \mathrm{MW/cm^2}$). Even a small amount of heat in the nonlinear cavity moves the modes and shifts the cavity to a regime where phase matching is not fulfilled.

Our experiments clearly show that we can fabricate high Q optical WG cavities from crystals using relatively simple mechanical polishing techniques. This manner of cavity preparation does not destroy the crystalline structure, nor does it affect the domain pattern that is important for quasiphase matching of the fundamental and second harmonics supported by WG modes in the cavity. Furthermore, domain boundaries do not induce any additional optical loss in periodically poled material, allowing a quality factor $Q = 2 \times 10^8$ to be achieved, both in PPLN and in congruent lithium niobate resonators.

In conclusion, we have shown that nonlinear optics with high Q WG cavities is important and interesting because it allows interaction of light with a material over effective length of tens and hundreds meters. It is practically impossible to fabricate a high quality stoichiometric crystal of this size. With WG mode cavities, however, surface reflection does the trick: For light circulating inside the millimeter-size cavity, the crystal appears to be very long. This opens novel opportunities for both fundamental science investigations and engineering applications of crystals. The example of the high efficiency nonlinear parametric process discussed above is only the beginning.

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