Highly Tunable Low-Threshold Optical Parametric Oscillation
in Radially Poled Whispering Gallery Resonators

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Whispering-gallery resonators (WGR's), based on total internal reflection, possess high quality factors in a broad spectral range. Thus, nonlinear-optical processes in such cavities are ideally suited for the generation of broadband or tunable electromagnetic radiation. Experimentally and theoretically, we investigate the tunability of optical parametric oscillation in a radially structured WGR made of lithium niobate. With a 1.04 μm pump wave, the signal and idler waves are tuned from 1.78 to 2.5 μm—including the point of degeneracy—by varying the temperature between 20 and 62 °C. A weak off centering of the radial domain structure extends considerably the tuning capabilities. The oscillation threshold lies in the mW-power range.

Optical micro-resonators (micro-cavities) attract increasing research interest owing to their outstanding properties and promises for numerous applications [1–4]. Ultrahigh quality factors of optical modes, reaching $Q \approx 10^{11}$ [5], together with small mode volumes provide unprecedented conditions for shaping and enhancement of light-matter interactions. Already existing and highly demanded applications of these fascinating new optical elements span from quantum electrodynamics and optics to optical filters and sensors [1,4,6].

Bringing nonlinear-optical effects to the range of low-power continuous-wave (cw) light sources is one of the biggest challenges. Whispering-gallery resonators (WGR's), made of crystalline nonlinear media, are best suited for this purpose [5,7,8]. The quality factors, which are restricted from above by light absorption, are among the highest ones. Together with a μm-size transversal confinement, a $\sim 10^7$ intensity enhancement can be reached. The discrete WGR mode structure is well understood nowadays and the techniques for its engineering and coupling light in and out are well developed [9–11].

For most of the nonlinear-optical phenomena, phase matching is a key issue. In WGR's it acquires specific features: While the most excitable equatorial and near-equatorial modes can be treated as plane waves, one has to take into account a discrete geometry-dependent mode structure as well as a modified dispersion relation [9]. The main impact of the discreteness is here in the reduced possibility to meet phase matching. The smaller the WGR size, the stronger is this impact [12]. Usually, the fine adjustment to narrow-band cavity nonlinear resonances can be made by varying the temperature or applying an electrical field [13].

A number of important nonlinear effects have been realized in WGR's with low-power cw light sources during the last years [2,3,13–20]. In $\chi^{(3)}$ optical materials, Raman scattering and lasing, third-harmonic generation, low-threshold optical oscillation owing to resonant four-wave mixing, and optical comb generation via Kerr nonlinearity were reported. In $\chi^{(2)}$ materials, second-harmonic generation, third-harmonic generation by cascading two second-order processes, and parametric down conversion were demonstrated recently.

Here we tackle the fundamental problem of tunability of nonlinear processes in WGR's by indicating how to vary optical frequencies over wide ranges. Specifically, we demonstrate for the first time a highly tunable low-threshold optical parametric oscillation (OPO) in a radially poled WGR made of lithium niobate (LiNbO$_3$).

As it is known from the studies of $\chi^{(2)}$ nonlinear-optical effects in bulk crystals, quasi-phase matching via domain engineering [21] provides great scope for tuning [22]. In the WGR case, a radial structure with an even number $2N \gg 1$ of domains, see Fig. 1(a), is best suited for that purpose. For the desired nonlinear process, the optimal large radius R at a specific domain number can be evaluated taking into account the geometry-dependent corrections to refractive indices of the interacting waves [10].

In polar optical materials, including lithium niobate, domain inversion results in changing the sign of the nonlinear susceptibility $\chi^{(2)}$. To account for the $2\pi R$ periodicity of the WGR case, we represent the sign-changing distribution $\chi^{(2)}(x)$, where x is the circumference coordinate, by the Fourier series

$$S(x) = \sum_{j=-\infty}^{\infty} S_j \exp\left(i\frac{jx}{R}\right), \qquad S_j = S_j^*, \qquad (1)$$
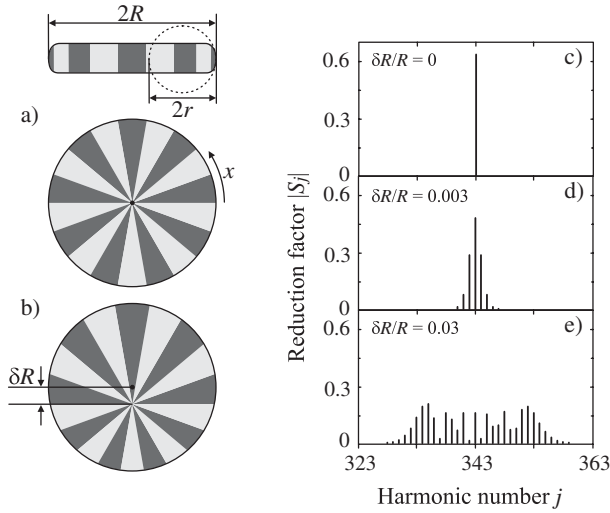
FIG. 1.   Geometric properties of radially poled WGR's: Ideal and distorted radial domain structures—(a) and (b)—and representative spectra of the reduction factor $|S_j|$—(c),(d), and (e)—for $2N = 686$ and $\delta R/R = 0$, 0.003, and 0.03, respectively, as defined by Eq. (1). Light and dark gray regions in (a) and (b) indicate the alternating domains, x is the circumference coordinate, R and r are the large and small WGR radii.

where the Fourier coefficients are given by the average over the circumference, $S_j = \langle S(x) \exp(-ijx/R) \rangle$, and $S(x) = \mathrm{sgn}[\chi^{(2)}(x)]$. The $1/R$ separation between the spatial frequencies, dictated by the $2\pi R$ periodicity, is indeed the same as the wave vector difference for the equatorial modes. As soon as the domain pattern (the points of the sign change) is known, one can calculate S_j numerically. The quantity $|S_j|$ represents the reduction factor for the nonlinear coefficient when using the j harmonic; it depends solely on the WGR geometry. If the domain pattern is strictly periodic, the Fourier components S_j can easily be calculated analytically. Only the harmonics with $j = \pm N, \pm 3N, \ldots$ are nonzero in this case, and the reduction factor for the main harmonic, $|S_N| = 2/\pi$, is close to 1. Any distortion of the periodicity of the domain structure makes nonzero all spatial harmonics S_j.

In our case, the most significant distortion originates from off-centering of the radial structure; see Fig. 1(b). It gives close side harmonics with $j = N \pm 1$, $N \pm 2$, etc. Figures 1(c)–1(e) illustrate the impact of off centering on the Fourier spectrum of the reduction factor $|S_j|$ for $2N = 686$, which is relevant to our experiment. One sees that the first side harmonics become comparable with the main one already for $\delta R/R = 0.003$. For $\delta R/R = 0.03$, we have a rich discrete spectrum, where the main harmonic is not the dominating one. The reduction factor remains pretty large for many spatial harmonics. Failure in the inversion of part of the domains is not critical for the spectrum, because the long-range order is maintained.

Each S_j peak can generally be used for phase matching. In the OPO case, the corresponding phase-matching conditions read

$$\frac{1}{\lambda_p} = \frac{1}{\lambda_s} + \frac{1}{\lambda_i}, \qquad \frac{n_p}{\lambda_p} = \frac{n_s}{\lambda_s} + \frac{n_i}{\lambda_i} + \frac{j}{2\pi R}, \qquad (2)$$

where $\lambda_{p,s,i}$ are the pump, signal, and idler wavelengths. The effective refractive index n, entering these relations, is a known function of λ, T, R, and r [10,23].

Figure 2 shows tuning curves calculated numerically from Eqs. (2) for a pump wavelength $\lambda_p = 1.04~\mu$m, the WGR radii $R = 1.54$ mm and $r = 0.6$ mm, extraordinary polarization for all three waves, and several values of j. Remarkably, there is a substantial difference in the tuning curves even for neighboring S_j-peaks. In essence, the tuning possibilities can be considerably extended in the non-ideal case when using several strongest spectral peaks instead of a single one. This extension occurs at the expense of a modest lowering of the nonlinear coupling strength leading to a modest increase in the threshold intensity.

According to Fig. 2, phase matching is possible only for $j \geq 342$ above room temperature. Closer examination shows that tuning by heating of the WGR becomes possible only owing to the impact of the off-centering if the large radius R exceeds noticeably the optimum value of 1.54 mm. Note lastly that the points of degeneracy, where $\lambda_s(T) = \lambda_i(T)$, are almost equidistant for the neighboring tuning curves with a temperature step $\Delta T \approx 22\,°$C.

In order to realize the extended OPO tunability, we have fabricated a radially poled WGR with $2N = 686$, $R \simeq 1.55$ mm, and $r \simeq 0.6$ mm. The corresponding R/r ratio is known to be favorable for coupling light in and out with a rutile (TiO$_2$) prism [11]. The resonator was fabricated from a 500 μm thick z-cut wafer of congruent lithium niobate. The domain structuring was performed using the standard electric-field-poling technique with liquid electrodes [24]; a radially patterned photo-resist corresponded to the desired domain number. The quality of the structuring was checked via domain selective etching—more than 50% of the desired domains are properly inverted. The radially poled part of the wafer was cut out and shaped (diamond-turned and polished) into a WGR with the
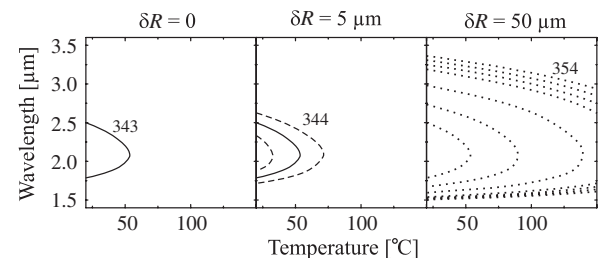

FIG. 2.   Tuning curves $\lambda_{s,i}(T)$ for $\lambda_p = 1.04~\mu$m, $2N = 686$, $R = 1.54$ mm, $r = 0.6$ mm, and $\delta R/R = 0$, 0.003, and 0.03. The solid lines correspond to the strongest spectral peaks with $|S_j| > 0.3$, the dashed lines to $0.2 < |S_j| \leq 0.3$, and the dotted lines to $0.1 < |S_j| \leq 0.2$. The curves corresponding to $|S_j| < 0.1$ are dropped.

desired R/r ratio. The off-center parameter δR was estimated to be within 50 μm leading to $\delta R/R \approx 0.03$.

Optical characterization of the fabricated WGR via line width and free-spectral-range measurements has shown that the intrinsic quality factor of the fundamental modes at $\lambda \simeq 1.04$ μm is $Q_p \approx 4 \times 10^7$ and the large radius $R \simeq 1.58$ mm. The last number is about 2.5% larger compared to the desired one (1.54 mm); the difference is within the accuracy of our fabrication procedure. Correspondingly, phase matching is expected for $j \geq 350$ instead of 342.

The setup for our optical experiments, depicted in Fig. 3, comprises the whispering-gallery resonator mounted on a heatable post. The latter one is used to change the resonator temperature up to $62\,^\circ$C. An extraordinarily polarized pump beam at 1.04 μm from an external-cavity diode laser is focused into a rutile prism. In the focus, placed at the prism base, the pump beam couples into the WGR via frustrated total internal reflection. In order to keep the setup as simple and as stable as possible, even under temperature expansion of the setup elements, the coupling prism contacts the resonator rim. The residual pump wave and the generated waves coupled out of the resonator are guided to a spectrometer covering the wavelength range from 0.2 to 1.1 μm. The spectra are collected for varying temperatures and pump powers.

As soon as the pump wave is coupled into the WGR, two narrow peaks corresponding to the pump frequency ν_p and to the second harmonic $2\nu_p$ become clearly visible in the spectrum. At pump powers P exceeding a threshold value $P_{\text{th}} \simeq 6$ mW, parametric oscillation has been detected. In addition to the former spectral features, new peaks arise well above the noise level, see Fig. 4. They can be reliably identified with the sum frequencies $\nu_p + \nu_s$ and $\nu_p + \nu_i$, where $\nu_{s,i}$ are the signal and idler frequencies, such that $\nu_s + \nu_i = \nu_p$. The accuracy of such measurements of $\nu_{i,s}$ is within the line width. In some cases, not only a single process, but also two parametric processes, corresponding to adjacent values of j, are observed simultaneously. While no direct measurements of the signal and idler waves were possible with our spectrometer, the data obtained give a complete picture of the actual nonlinear processes.

Changing the temperature T between 20 and 62 $^\circ$C, we were able to tune the signal and idler wavelengths from 1.78 to 2.5 μm, including the point of degeneracy $\lambda_s = \lambda_i$, in steps of (1–2) nm. The corresponding experimental data are presented by the dots in Fig. 5. Most of the experimental dots nicely follow three tuning curves (solid lines) calculated for the neighboring spectral peaks with $j = 350$, 351, and 352. In accordance with theory, the measured temperature difference between two adjacent points of degeneracy is about 22 $^\circ$C. Remarkably, filling of different tuning curves with the dots is nonuniform. For $T \lesssim 30\,^\circ$C, the curve with $j = 352$ is practically unfilled. The branches with $j > 352$ remain inactive in the whole temperature range.

Several issues, closely related to the above results, are worthy of discussion:

In some special cases, phase matching for $\chi^{(2)}$ nonlinear processes can be realized in WGR's even without domain engineering, i.e., in the single-crystal case. With lithium niobate, it is possible for the processes involving modes of different polarizations [13,20]. However, the tunability is restricted and the relevant nonlinear coefficient d_{13} is about 5 times smaller than the largest coefficient d_{33} [25].

Being focused on the tunability issue, we did not try to minimize the pump threshold P_{th}. While our experimental value $P_{\text{th}} \approx 6$ mW is already low, it can still be decreased considerably. What are the prospects for decreasing the threshold power? In order to estimate them, we write down the scaling relation $P_{\text{th}} \propto \nu_i \nu_s / Q_p^* Q_s^* Q_i^* d_j^2$, where $d_j = |S_j| d_{33}$ is the effective nonlinear coefficient and $Q_{p,s,i}^*$ are the loaded quality factors incorporating the coupling losses [26]. This relation contains the main parameters which can be varied in the experiment. Because of the imperfect domain engineering, see above, the actual reduction factors $|S_j| \approx 0.1$ were (3–4) times smaller than they might be, which gives roughly 1 order of magnitude increase in P_{th}.

FIG. 4 (color). Spectrum of the visible output of the WGR at a pump power of 10 mW for two different temperature values showing sum frequency peaks at $\nu_p + \nu_s$ and $\nu_p + \nu_i$. The corresponding parametric processes are phase matched at $j = 351$ and 352.
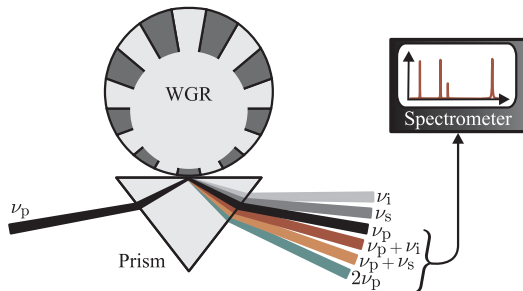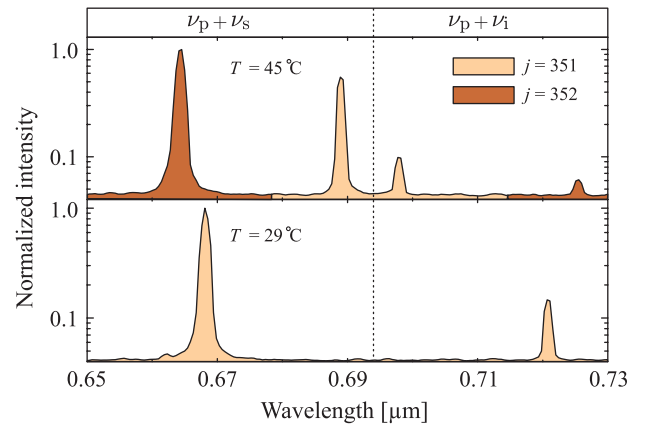

FIG. 3 (color). Schematic of the WGR-OPO experimental setup. The frequencies of the interacting waves are ν_p (pump), ν_s (signal), and ν_i (idler).
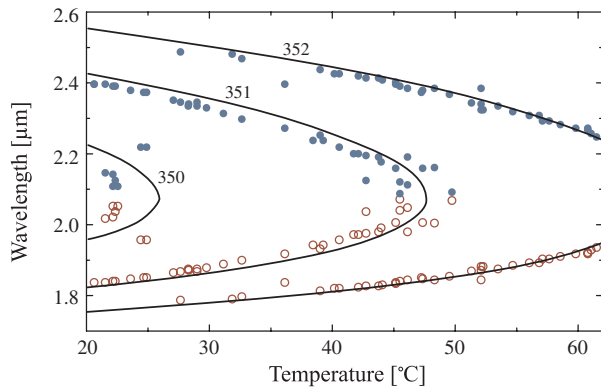
FIG. 5 (color).   Signal and idler wavelengths (open and filled dots) evaluated from the frequency spectra as functions of the temperature T. The solid lines are the tuning curves calculated numerically for $j = 350$, 351, and 352 from Eq. (2).

About 2 orders of magnitude increase stems from the overall coupling losses for p, s, and i waves—bringing the coupling prism in contact with the WGR rim noticeably decreases $Q^*_{p,s,i}$. Thus, the threshold power can be decreased by about 3 orders of magnitude without sacrificing the tuning properties.

It is known that the phase-matching acceptance bandwidth increases nearby the point of degeneracy, $\lambda_s = \lambda_i$, if the s, i modes are of the same polarization [27]. This is why the experimental dots in Fig. 5 are widely spread around the tuning curve for $j = 351$ at $T \approx 45\,°C$. If a narrow acceptance bandwidth is required, one can employ an OPO scheme with different s, i polarizations. The domain number $2N$ must be different in this case. About 1480 domains are needed, instead of former $2N \approx 700$, to realize this scheme for WGR radii similar to the ones in our experiment. The structure of the tuning curves is expected to be different as well. The above mentioned indicates a high degree of flexibility of the nonlinear schemes based on structured WGR's. By changing the domain number one can proceed from broadband to narrow-band gain and from identically polarized to cross-polarized generated waves.

In conclusion, we have shown that radial poling of whispering-gallery resonators made of lithium niobate allows us to combine a high tunability of nonlinear-optical processes, such as optical parametric oscillation, with common low-power continuous-wave light sources. The tuning characteristics differ drastically from those known for bulk nonlinear schemes because of the discrete geometry-dependent mode structure. The potential of quasi-phase-matching for shaping nonlinear processes is thus strongly extended.

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