Low-threshold parametric nonlinear optics with quasi-phase-matched whispering-gallery modes

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We propose to fabricate a toroidal dielectric cavity from a periodically poled $\chi^{(2)}$ nonlinear material (e.g., $LiNbO_3$) to achieve efficient interaction among high-Q whispering-gallery modes. We show that the periodic poling allows for suppression of both material and cavity dispersion. Such a cavity might be a basic element of a family of efficient nonlinear devices operating at a broad range of optical wavelengths. © 2003 Optical Society of America

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1. INTRODUCTION

Whispering-gallery modes (WGMs) in optical microcavities are attractive in nonlinear optics because of their small mode volumes and high quality.[1,2] Efficient parametric nonlinear interaction among modes is possible if the cavity that supports WGMs is fabricated from a low-loss $\chi^{(2)}$ nonlinear material. However, nonlinear parametric interactions are usually strongly forbidden by the law of conservation of momentum (phase matching condition) because WGMs of a dielectric cavity that possess rotational symmetry are orthogonal to one another in momentum space. Parametric interaction occurs if the symmetry of the system is broken or modified.

A solution to this problem was recently suggested for strongly nondegenerate three-wave mixing.[3,4] Efficient resonant interaction of light confined in three optical WGMs and a microwave field was achieved by engineering of the geometry of a microwave resonator coupled with a toroidal $LiNbO_3$ optical cavity. A new kind of electro-optic modulator–photonic receiver based on this interaction was suggested and achieved.[3–8]

Here we propose an approach to achieving parametric interaction among different WGMs by use of a toroidal cavity produced from a periodically poled nonlinear material.[9] As an example, we calculate the structure of the cavity poling that is necessary for creation of a triply resonant optical parametric oscillator that converts a pump photon ($\lambda_0 = 1064$ nm) into signal and idler photons ($\lambda_s = \lambda_i = 2128$ nm). Such a parametric oscillator might have submicrowatt threshold power for realistic parameters of the system.

Our results are promising for various applications in nonlinear and quantum optics that range from optical frequency conversion, optical modulation, and photonic reception to production of nonclassical states of light, quantum nondemolition measurements, and quantum computing.

2. PHASE MATCHING OF WHISPERING-GALLERY MODES

The problem of creating efficient wave mixing with whispering-gallery modes stems from two sources: frequency-dependent dispersion of the host material of the dielectric cavity and dispersion introduced by the internal geometrical mode structure. WGMs of high order may be understood as dielectric optical waveguides. WGMs have dispersion that depends on geometry, similarly to waveguides. The frequency of a high-order TE WGMs may be estimated from the equation

$$\frac{2\pi R}{\lambda}\sqrt{\epsilon(\lambda)} + \left[\frac{\epsilon(\lambda)}{\epsilon(\lambda) - 1}\right]^{1/2} = \nu + \alpha_q\left(\frac{\nu}{2}\right)^{1/3} + \frac{3\alpha_q^{\,2}}{20}\left(\frac{2}{\nu}\right)^{1/3} + O(\nu^{-2/3}), \quad (1)$$

where λ is the wavelength in vacuum; ν is the mode order; $\epsilon(\lambda)$ is the susceptibility of the nonlinear material; R is the radius of the cavity; and α_q is the qth root of Airy function $\mathrm{Ai}(-z)$, which is 2.338, 4.088, and 5.521 for $q = 1, 2, 3$, respectively.

Equation (1) was derived under the assumption that the cavity is a central part of a dielectric sphere or spheroid. Because the main sequence of WGMs is localized close to the sphere equator it is possible to remove an unused part of the sphere to create a disk cavity without changing the mode spectrum. The angle distribution of the field is determined by $\mathbf{Y}_{\nu,m}$ spherical functions with angular number ν and magnetic number m. For the main sequence of WGMs we take $m = \nu$. The radial WGM profile for a spherical cavity is $\Psi(r) = J_{\nu+1/2}(kr)/r$, where $J_{\nu+1/2}(kr)$ is a Bessel function of the first kind and $\nu = 0, 1, 2, 3, \ldots$. The mode spectrum results from the boundary condition $\Psi(r) \to 0$ for $r \to \infty$ and $r \to 0$. Equation (1) is valid for WGMs characterized by $\nu \gg 1$.[10]

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Cutting a crystal sphere is not practical. One may fabricate a cavity by polishing the rim of a dielectric cylinder cut from a crystal wafer.[7,8] The rim of such a cavity would not necessarily need to be spherical to maintain high-Q and small volume WGMs. It was shown experimentally that the rim may be spheroidal or even conical.[11] Calculation of the spectrum of an arbitrary dielectric spheroid is rather complicated. However, Eq. (1) is still valid for the main mode sequence.[8,12]

We assume that the cavity is fabricated from a commercial flat Z-cut $LiNbO_3$ substrate, so TE modes correspond to the extraordinary waves in the material. For example, let us consider a degenerate optical parametric oscillator (OPO) pumped at $\lambda_p = 1064$ nm. Susceptibilities for pump, signal, and idler waves are $\epsilon(\omega_p) = 4.657$ and $\epsilon(\omega_s) = \epsilon(\omega_i) = 4.513$, respectively.

To achieve phase matching for such an OPO in a bulk, congruent $LiNbO_3$, poling periods Λ have to obey the following condition:[9]

$$k_p - k_s - k_i - \frac{2\pi}{\Lambda} = 0, \tag{2}$$

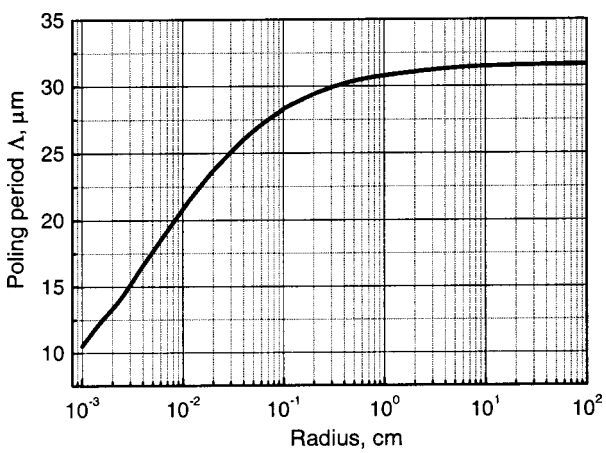

Fig. 1. Dependence of poling period Λ on cavity radius. It is easy to see that for a large cavity radius the period coincides with the period in a bulk medium, whereas for a smaller radius the period decreases.

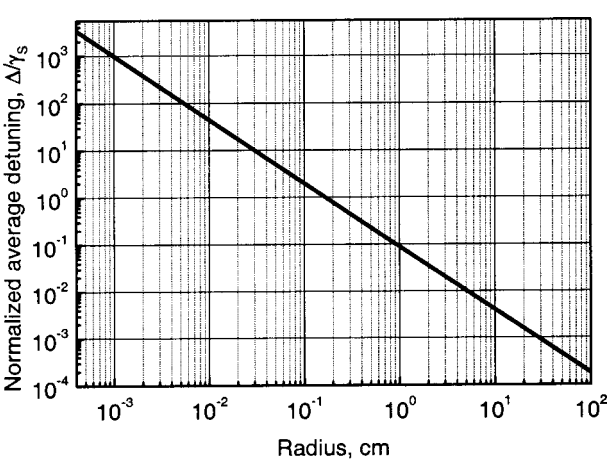

Fig. 2. Average detuning of the signal frequency from the nearest WGM of a dielectric cavity versus cavity radius. The pump field ($\lambda_p = 1.064$ μm) is resonant with a mode of the $LiNbO_3$ cavity.

where k_p, k_s, and k_i are the wave vectors of the pump, the signal, and the idler, respectively. For the degenerate case that we consider here, the poling period is determined by

$$\Lambda = \frac{\lambda_p}{\sqrt{\epsilon_p} - \sqrt{\epsilon_s}} \approx 31.67\ \mu\text{m}. \tag{3}$$

For a WGM cavity fabricated from the same material the poling period is different because of the dependence of the mode dispersion on the cavity's geometrical parameters, as will be shown below. The smaller the cavity, the shorter the period (see Fig. 1). Moreover, there is no guarantee that the pump and the signal frequencies are both resonant with the cavity modes. To characterize this possibility we assume that the pump is resonant with a cavity mode and look for the detuning

$$\frac{\Delta}{\gamma_s} = \left|\frac{\omega_p}{\tilde{\omega}_s} - 2\right| Q_s, \tag{4}$$

where γ_s and Q_s are the half-linewidth and the quality factor of the signal mode, $Q_s = \omega_s/(2\gamma_s)$; $\tilde{\omega}_s$ is the frequency of the cavity mode that minimizes Δ. If $\Delta/\gamma_s < 1$ we may say that the signal frequency is resonant with the cavity mode. The dependence of the detuning on the cavity radius in not simple. The average dependence is presented in Fig. 2 for $Q_s = 10^7$. We may conclude from this calculation that the smaller the cavity, the less the probability that it has both resonant frequencies ω_p and $\omega_p/2$. As follows from Fig. 2, the cavity radius should exceed 2 mm for this condition to be fulfilled.

However, the condition $\Delta/\gamma_s < 1$ is not critical even for smaller cavities. The frequency difference may be compensated for by application of a dc bias field to the cavity. The dc field moves modes of different frequencies in a different way, which results in the frequency matching in the system.

Let us consider a cavity with radius $R = 0.64$ mm and study the main mode sequence ($\alpha_q = 2.338$). It is worth noting that mechanical preparation of high-Q cavities with such a radius for millimeter-wave electro-optical modulation was recently demonstrated.[8] The mode that is resonant with the pump field has index $\nu_p = 8156$. For the signal and idler modes, $\nu_s = \nu_i = 4004$. Because $\nu_p - \nu_s - \nu_i = 148 > 1$ there is no parametric interaction in the system unless the nonlinear index of refraction of the cavity material is modulated with the period determined by the number $\nu_\chi = \nu_p - \nu_s - \nu_i$. Modulation with period ν_χ is indeed the basis for Fig. 1, because ν_χ/ν_s increases as R decreases. This periodicity also results in phase matching for degenerate parametric frequency downconversion with pump radiation of 1.056, 1.049, 1.041 μm, and so on.

The approximate modulation period is 27.3 μm. It is easy to achieve such modulation by periodic poling of the cavity material.[9] The volumes of the pump mode, the signal–idler mode, and modes' overlap are 1.7×10^{-7}, 4×10^{-7}, and 1.5×10^{-7} cm^3, respectively (see the mode profiles in Fig. 3). Note that the mode volumes can also be estimated by use of the asymptotic expression $V_p \approx 2\pi R \times 2R(2\pi/\nu_p)^{1/2} \times (R/\nu_p^{2/3})$, where $2\pi R$ is

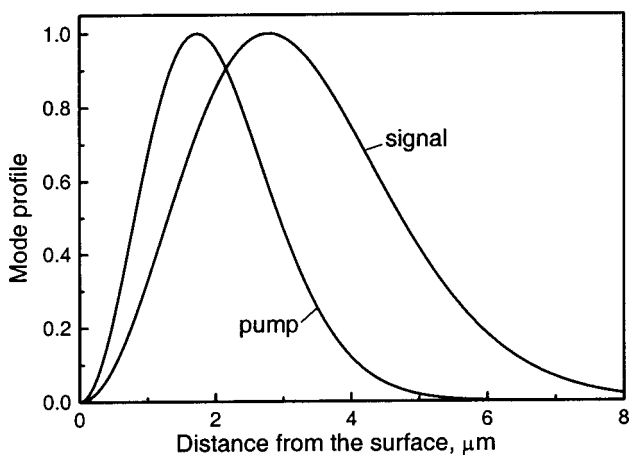

Fig. 3. Profiles of the field distribution inside a dielectric cavity. Zero coordinate corresponds to the cavity boundary. In our model we neglect the evanescent field outside the cavity, which is a reasonable approximation for high-Q WGMs.

the length of the cavity rim, $2R(2\pi/\nu_p)^{1/2}$ is the mode size in the Z direction, and $R/\nu_p^{2/3}$ is the radial size of the mode.

The radius of the cavity is contained only in the ratio R/λ in Eq. (1). Therefore the phase matching established by the periodic poling of the cavity material is stable with respect to the radius change, and the radius change may be compensated for by fine tuning of the pump laser frequency.

3. PARAMETRIC OSCILLATIONS IN A WHISPERING-GALLERY MODE CAVITY

Let us now estimate the threshold for parametric oscillation. The interaction energy between the pump and the signal modes may be written as

$$\mathcal{V} = \int_V \chi^{(2)} E_p E_s^2 \mathrm{d}V, \tag{5}$$

where $\chi^{(2)}$ varies in the space nonlinearity of the cavity, E_p and E_s are the pump and the signal mode amplitudes, respectively (the signal mode coincides with the idler mode), and V is the volume of the cavity. We present mode amplitudes as

$$E_p = \left(\frac{2\pi\hbar\omega_p}{\epsilon_p V_p}\right)^{1/2} \Psi_p(r)\exp(i\nu_p\phi)a_p\exp(-i\omega_p t) + \text{adjoint}, \tag{6}$$

$$E_s = \left(\frac{2\pi\hbar\omega_s}{\epsilon_s V_s}\right)^{1/2} \Psi_s(r)\exp(i\nu_s\phi)a_s\exp(-i\omega_s t) + \text{adjoint}, \tag{7}$$

where a_i and $a_i^\dagger$ $(i = p, s)$ are annihilation and creation operators for the mode, $\Psi_p(r)$ and $\Psi_s(r)$ are the mode spatial profiles normalized such that $V_i = \int_V |\Psi_i(r)|^2 \mathrm{d}V$ $(i = p, s)$, ω_p and ω_s are the mode frequencies, and ϵ_p and ϵ_s are the susceptibilities of the material, respectively. The problem of quantization of electromagnetic waves in dielectrics was discussed in Refs. 13 and 14. We also assume that the nonlinearity of the medium is modulated such that it has a Fourier component that matches the signal and pump modes: $\chi^{(2)} \to 2\tilde{\chi}^{(2)}\cos[(\nu_p - 2\nu_s)\phi]$.

We now write the interaction Hamiltonian in the slowly varying amplitude and phase approximation:

$$H = \hbar g[(a_s^\dagger)^2 a_p + a_p^\dagger a_s^2], \tag{8}$$

where the coupling constant is

$$g = 2\pi\omega_s \frac{\tilde{\chi}^{(2)}}{\epsilon_s}\frac{V_{pss}}{V_s}\left(\frac{2\pi\hbar\omega_p}{\epsilon_p V_p}\right)^{1/2}, \tag{9}$$

$V_{pss} = \int_V \Psi_p \Psi_s^2 \mathrm{d}V < V_p$, and V_s is the mode overlap integral.

Using this Hamiltonian, we derive equations of motion:

$$\dot{a}_p = -\gamma_p a_p - i g a_s^2 + F_p, \tag{10}$$

$$\dot{a}_s = -\gamma_s a_s - 2ig a_s^\dagger a_p + F_s, \tag{11}$$

where F_p and F_s are the Langevin forces and γ_p and γ_s are pump and signal decay rates, respectively. The expectation value $\langle F_p\rangle$ describes pumping from outside the system. We can write the expression $|F_p|^2/\gamma_p^2 = 4W_p Q_p/(\hbar\omega_p^2)$,[8] where $Q_p = \omega_p/(2\gamma_p)$ is the mode quality factor and W_p is the power of the pump radiation.

Solving Eqs. (10) and (11) in steady state and neglecting quantum fluctuations, we find the expectation value of the photon number for the signal that has been generated:

$$|a_s|^2 = \frac{\gamma_p\gamma_s}{2g^2}\left(\frac{2g}{\gamma_s\gamma_p}|\langle F_p\rangle| - 1\right), \tag{12}$$

which results in the threshold condition for parametric oscillation:

$$W_p \geqslant \frac{\epsilon_p \epsilon_s^2}{512\pi^3[\tilde{\chi}^{(2)}]^2}\left(\frac{V_s}{V_{pss}}\right)^2 \frac{\omega_p V_p}{Q_s^2 Q_p}. \tag{13}$$

For realistic parameters ($V_{pss}/V_s = 0.5$, $V_p = 2 \times 10^{-7}$ cm^3, $\epsilon_p = 4.6$, $\epsilon_s = 4.5$, $\tilde{\chi}^{(2)} = 10^{-7}$ cgs, $\omega_p = 2 \times 10^{15}$ s^{-1}, $Q_s \simeq Q_p \simeq 10^7$) the threshold value is 0.1 μW, which is orders of magnitude less than the state-of-the-art OPO threshold power, e.g., 0.5 mW for similar wavelengths.[15]

We believe that the toroidal cavity is better than the total-internal-reflection cavity used in OPOs pumped at 1064 nm.[16] The oscillation threshold depends on the mode volume and the mode overlap integral. We can reduce the mode volume and increase the integral in WGM cavity.

4. DISCUSSION

Let us discuss the possibilities of fabricating nonlinear cavities. The optimum poling geometry is symmetric with respect to the center of the cavity [Fig. 4(a)]. When the nonlinear index coefficient is modulated with a periodic sign reversal, the Fourier coefficient for the first (fundamental) harmonic is $\sim 2/\pi$, as in a periodically poled bulk material. The dependence of the Fourier coefficients on the inverse poling period is shown in Fig. 5,

where we average the discrete spectrum and show its envelope function.

Fabrication of centrally symmetric poling is a difficult task. It seems to be much easier to use a slice of commercially available periodically poled $LiNbO_3$. The cavity will have poled stripes instead of sectors [see Fig. 4(b)]. For such poling, a wave that travels close to the cavity surface sees a nonlinear grating with a changing period. An envelope function for the Fourier decomposition for such a grating is shown in Fig. 6. Because the grating does not have a fixed step with respect to the electromagnetic wave, the maximum Fourier component is less than in the ideal case (symmetric poling), which leads to an increase in the oscillation threshold compared with that for periodic poling (11 times in the case considered). However, the spectrum of the grating is broad enough to simplify working with the cavity and to make possible multifrequency parametric oscillations in the system.

Nonequidistant poling stripes may, in principle, create more-periodic poling and change the spectrum. However, cutting a disk from a sample with complicated poling structure is not an easy task.

It is worth mentioning that the nonlinear system discussed above may also be used to produce nondegenerate parametric interactions. Because of the low threshold the system may also be reversed and used as a detector of long-wavelength radiation by means of its direct upconversion into light. The maximum wavelength of the radiation is determined by the cavity size. To maintain low radiative losses one should make the order of the cavity mode sufficiently large. For example, for detection of 10-μm radiation the cavity should have a radius of at least 3.5 mm.

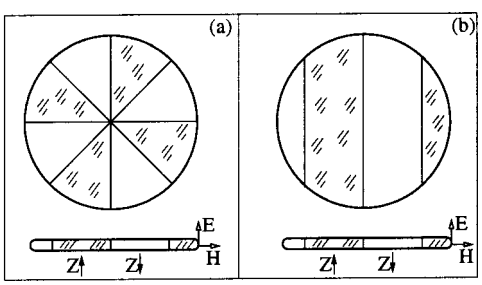

Fig. 4. Examples of material poling to achieve phase matching for WGMs: (a) poling symmetric with respect to the cavity center, (b) stripelike poling.

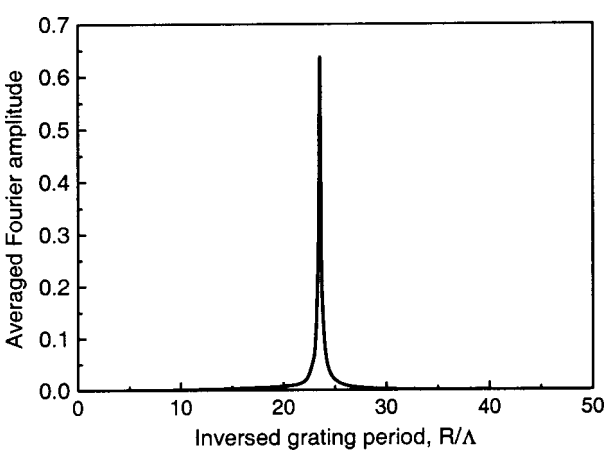

Fig. 5. Envelope function for the amplitudes of Fourier coefficients for the poling shown in Fig. 4(a).

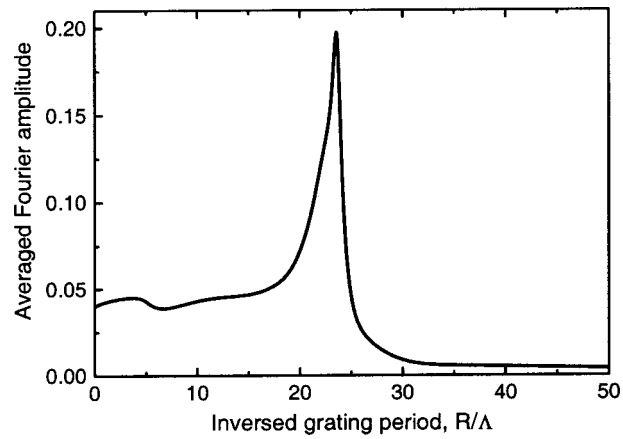

Fig. 6. Envelope function for the amplitudes of Fourier coefficients for the poling shown in Fig. 4(b).

Moreover, periodically poled materials may be used for construction of electro-optic modulators based on WGM, instead of the specially shaped microwave cavity that was utilized for efficient modulation of light in previous studies.[6,8] The usefulness of periodically poled materials for modulation of light was recognized more than a decade ago[17] and further developed.[18–21] Possibilities of parametric interaction among running light-wave and microwave radiation in a periodically poled WGM cavity as well as single-sideband electro-optical modulation by use of nonequidistant modes of a WGM cavity are discussed in Ref. 22.

5. CONCLUSIONS

We have proposed fabrication of a whispering-gallery-mode dielectric cavity from a periodically poled nonlinear material. We have shown that appropriate periodic poling leads to compensation for the phase mismatch among cavity modes that results from cavity material dispersion as well as from geometrical cavity dispersion. We showed that because of the potential high-quality factors and small volumes of the modes one can use them to create an ultralow-threshold optical parametric oscillator.

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