# Second Order Parametric Processes in Nonlinear Silica Microspheres

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We analyze second order parametric processes in a silica microsphere coated with radially aligned nonlinear optical molecules. In a high-$Q$ nonlinear microsphere, we discover that it is possible to achieve ultralow threshold parametric oscillation that obeys the rule of angular momentum conservation. Based on symmetry considerations, one can also implement parametric processes that naturally generate quantum entangled photon pairs. Practical issues regarding implementation of the nonlinear microsphere are also discussed.

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Because of its unrivaled low propagation loss, silica glass has become a very important medium for the study of nonlinear optics [1]. However, under the standard dipole approximation, bulk silica glass, being centrosymmetric, cannot possess any second order nonlinearity. Consequently, investigation of nonlinear optics in silica-based materials focuses primarily on third order processes such as Kerr effects. In a recent Letter [2], we demonstrated that a silica fiber coated with radially aligned nonlinear molecules, despite its highly symmetric geometry, can exhibit large and thermodynamically stable second order nonlinearity. Both theoretically and experimentally, evidence that centrosymmetry does not preclude second order nonlinear response has been reported in the context of colloidal particles in bulk solution [3–5]. Existing work in this area, however, focuses on particles with a relatively small size, with dimension of the order of 1 $\mu$m or less. Therefore, to the best of our knowledge, the possibility of using high-quality factor whispering gallery (WG) modes in a microsphere [6–8] to generate strong second order nonlinear responses has not been explored. In this Letter, we investigate second order parametric processes within a nonlinear microsphere, which consists of a silica microsphere coated with radially aligned nonlinear molecules, as illustrated in Fig. 1(a). Because of the high-$Q$ WG modes and their symmetry properties, we find that it is possible to demonstrate ultralow threshold parametric oscillation and achieve symmetry-enforced quantum entanglement.

The WG modes in a microsphere can be classified as either transverse electric (TE) or transverse magnetic (TM). For a TM mode, by using Eqs. (9.118) and (10.60) in Ref. [9], we can express its electric field as:

$$\vec{E}_{qlm} = -\frac{Z\sqrt{l(l+1)}}{kr} f_l(r) Y_{lm} \hat{e}_r + \frac{iZ}{kr}\frac{\partial}{\partial r}[r f_l(r)] \hat{e}_r \times \vec{X}_{lm}, \tag{1}$$

where $\hat{e}_r$ is the radial unit vector, $Z$ is the wave impedance, $k$ is the dielectric wave vector, $f_l(r)$ is a linear combination of the $l$th order spherical Bessel functions, and $\vec{X}_{lm} = \vec{r} \times \vec{\nabla} Y_{lm}(\theta, \varphi)/i\sqrt{l(l+1)}$, with $Y_{lm}(\theta, \varphi)$ being spherical harmonics. The subscript $q$ is the radial quantum number and refers to the number of radial nodes in $f_l(r)$. The magnetic field of the WG mode in Eq. (1) is given by $\vec{H}_{qlm} = f_l(r)\vec{X}_{lm}(\theta, \varphi)$ [9]. We use the normalization of $\iiint |\vec{H}_{qlm}|^2 r^2 dr d\Omega = R^3$, which is equivalent to $\int |f_l(r)|^2 r^2 dr = R^3$, with $f_l(r)$ dimensionless. We denote the WG mode in Eq. (1) as $|q, l, m\rangle$.

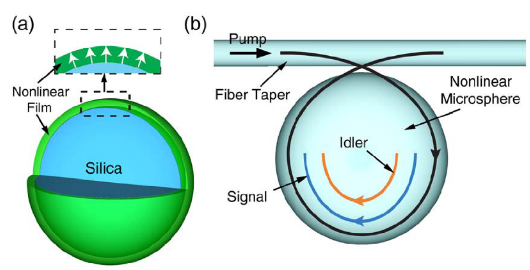


FIG. 1 (color online). (a) Schematic of a nonlinear microsphere. (b) A nonlinear microsphere coupled to a fiber taper. A pump high-$Q$ WG mode can be excited through taper-microsphere coupling. The signal and idler photons can be coupled out by using the same fiber taper or through an additional fiber taper or optical prism.

 

We now consider a nonlinear microsphere coupled to a fiber taper, as shown in Fig. 1(b). By carefully controlling the distance between the fiber taper and the microsphere, it is possible to convert almost all optical power in the fiber taper into a single high-$Q$ WG mode circulating within the microsphere [10]. This capability suggests that, through the fiber taper, we can couple pump photons at frequency $\omega_3$ into a high-$Q$ WG mode $|q_3, l_3, m_3\rangle$. Through nonlinear molecules, the pump photons can be split into signal photons at $\omega_1$ and idler photons at $\omega_2$, where $\omega_3 = \omega_1 + \omega_2$. We denote the electric field of the pump mode as $\vec{\mathbf{E}}_3 = \mathrm{Re}\{A_3 \exp[i\omega_3 t]\vec{E}_{q_3 l_3 m_3}(\vec{r})\}$, where $\vec{E}_{q_3 l_3 m_3}(\vec{r})$ is defined in Eq. (1) and $A_3$ is the mode amplitude. The signal and the idler fields can be similarly described by using subscripts 1 and 2. Since WG modes have finite linewidth, the photon frequencies may deviate slightly from the resonant peaks of WG modes. Consequently, for signal photons, we use $\omega_1$ to denote photon frequency and $\omega_{1,0}$ to represent the resonant frequency of the WG mode. Similar notations, labeled with subscripts 2 and 3, are used for idler and pump photons, respectively.

Second order parametric processes in the microsphere are governed by the nonlinear Maxwell's equation: $\nabla \times (\nabla \times \vec{E}) + \mu\varepsilon\partial^2\vec{E}/\partial t^2 = -\mu\partial^2\vec{P}_{\mathrm{NL}}/\partial t^2$ [11], where the nonlinear polarization $\vec{P}_{\mathrm{NL}} = \varepsilon_0\chi^{(2)} : \vec{E}\vec{E}$ is responsible for the coupling between the three parametric components. By assuming signal, idler, and pump photons in the WG modes of $|q_1, l_1, m_1\rangle$, $|q_2, l_2, m_2\rangle$, and $|q_3, l_3, m_3\rangle$ and applying the nonlinear Maxwell's equation, we find the following equations for mode amplitudes $A_1$, $A_2$, and $A_3$:

$$\frac{dA_1}{dt} = i(\omega_{1,0} - \omega_1)A_1 - \frac{1}{2}\frac{\omega_1}{Q_1}A_1 - \frac{i}{2}\omega_1\kappa A_3 A_2^*, \tag{2a}$$

$$\frac{dA_2}{dt} = i(\omega_{2,0} - \omega_2)A_2 - \frac{1}{2}\frac{\omega_2}{Q_2}A_2 - \frac{i}{2}\omega_2\kappa A_3 A_1^*, \tag{2b}$$

$$\kappa = \frac{\varepsilon_0}{\mu_0 R^3}\iiint \vec{E}^*_{q_1 l_1 m_1} : \chi^{(2)} : \vec{E}^*_{q_2 l_2 m_2}\vec{E}_{q_3 l_3 m_3} dv, \tag{2c}$$

where $Q_i$ is the modal $Q$ factor and $\kappa$ represents the strength of nonlinear coupling. [In deriving Eq. (2), we assume that the idler wave is spontaneously generated.] Parametric oscillation occurs under the condition of $dA_1/dt = dA_2/dt = 0$, which leads to $\omega_2(\omega_{1,0} - \omega_1)/Q_2 = \omega_1(\omega_{2,0} - \omega_2)/Q_1$ and $|\kappa|^2|A_3|^2 = \frac{1}{Q_1 Q_2} + \frac{4(\omega_{1,0}-\omega_1)(\omega_{2,0}-\omega_2)}{\omega_1\omega_2}$. A particularly simple yet physically significant solution is the on-resonance case, where we have $\omega_1 = \omega_{1,0}$, $\omega_2 = \omega_{2,0}$, and $\omega_3 = \omega_{1,0} + \omega_{2,0}$. Under the on-resonance condition, the pump threshold for parametric oscillation reaches its lowest value and is determined by

$$|A_3| = \frac{1}{|\kappa|\sqrt{Q_1 Q_2}}. \tag{3}$$

Since an all-silica microsphere can possess an extremely high-quality factor [6,7], Eq. (3) indicates that we need only very low pump power to achieve parametric oscillation.

To simplify Eq. (2c), we assume that the $\chi^{(2)}$ tensor is dominated by its radial component $\chi_{rrr}$. This is consistent with the configuration of radially aligned nonlinear molecules grown conformally on the microsphere surface [12]. This assumption also implies that we need only to consider the $E_r$ component of the TM modes and ignore any TE modes, since TE modes have no $E_r$ component. Furthermore, we assume that the nonlinear coating thickness $\delta$ is small and its index is similar to that of silica glass. This allows us to approximate the $E_r$ field in the nonlinear film as that of the WG mode at the microsphere surface. With these considerations, Eq. (2c) becomes

$$\kappa = -\Gamma^{l_3 m_3}_{l_1 m_1; l_2 m_2}\frac{\varepsilon_0 Z^3}{\mu_0 k_1 k_2 k_3 R^4}\delta\chi^{(2)}_{rrr} f^*_{l_1} f^*_{l_2} f_{l_3}, \tag{4}$$

where the impedance $Z$ and the wave vectors refer to the values in silica glass. In Eq. (4), $\Gamma^{l_3 m_3}_{l_1 m_1; l_2 m_2}$ is a dimensionless number that contains two parts, i.e., $\Gamma^{l_3 m_3}_{l_1 m_1; l_2 m_2} = \Gamma_1\Gamma_2$. $\Gamma_1$ is simply $\sqrt{l_1(l_1+1)l_2(l_2+1)l_3(l_3+1)}$. $\Gamma_2$ is given by $\iint Y^*_{l_1 m_1}Y^*_{l_2 m_2}Y_{l_3 m_3}d\Omega$, which is $\sqrt{(2l_1+1)(2l_2+1)/4\pi(2l_3+1)}\langle l_1 l_2; 00|l_1 l_2; l_3 0\rangle \times \langle l_1 l_2; m_1 m_2|l_1 l_2; l_3 m_3\rangle$. [See Eq. (3.7.73) of Ref. [13].] Of particular significance is the appearance of the Clebsch-Gordan coefficient $\langle l_1 l_2; m_1 m_2|l_1 l_2; l_3 m_3\rangle$. Taken together, Eq. (4) means that any second order nonlinear processes within the microsphere must obey the rule of angular momentum conservation: If the signal, idler, and pump photons are, respectively, in the states of $|q_1, l_1, m_1\rangle$, $|q_2, l_2, m_2\rangle$, and $|q_3, l_3, m_3\rangle$, they must satisfy $m_3 = m_1 + m_2$ and $|l_1 - l_2| \leq l_3 \leq l_1 + l_2$. Additionally, to satisfy the on-resonance condition, we require the frequency mismatch between the three WG modes to be less than the sum of their individual linewidths, i.e., $\Delta\omega \equiv |\omega_{3,0} - \omega_{1,0} - \omega_{2,0}| \leq \sum_{i=1}^{3}\omega_{i,0}/Q_i$. By assuming pump photons in the visible range and all $Q$ factors in the range of $10^6$, this on-resonance condition is equivalent to requiring the frequency detuning between the WG modes ($\Delta\nu \equiv \Delta\omega/2\pi$) to be less than 1 GHz. Since silica microspheres possess a large number of high-$Q$ WG modes, satisfying both the on-resonance condition and the requirement of angular momentum conservation is not difficult: For a microsphere with a radius of 15 $\mu$m and in the wavelength range of 200 nm to 3 $\mu$m, there are at least 62 sets of WG modes (excluding any degeneracy factors) that satisfy both requirements. In our calculations, the WG mode frequencies are obtained by using the field-matching technique in Ref. [14]. The dispersion of silica glass is fully taken into account by using the formula on page 8 of Ref. [1]. A specific example of frequency-matched WG modes is: $q_1 = 1$, $l_1 = 110$, $\lambda_1 = 1148.6331$ nm (signal); $q_2 = 2$,

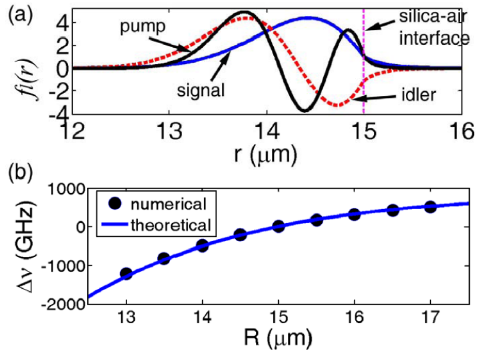


FIG. 2 (color online). (a) The normalized mode functions of three on-resonant high-$Q$ WG modes. The microsphere radius is 15 $\mu$m. (b) The frequency detuning between the three parametric components in (a) as a function of the microsphere radius $R$. The numerical and the theoretical results are obtained through field-matching calculations and perturbation analysis in Ref. [17], respectively.

$l_2 = 138$, $\lambda_2 = 882.3905$ nm (idler); $q_3 = 3$, $l_3 = 248$, $\lambda_3 = 499.03014$ nm (pump). The frequency detuning between the three modes is 562 MHz. The normalized mode functions $f_l(r)$ of all three WG modes are given in Fig. 2(a). With $l_3 = l_1 + l_2$, we can choose $m = l$ for all three WG modes to satisfy angular momentum conservation. The pump mode with $m = l$ can be easily excited by using the configuration in Fig. 1(b) [15].

We can combine Eqs. (1), (3), and (4) to estimate the electric field strength required for parametric oscillation. Equations (3) and (4) can give us the threshold value for the pump mode amplitude $A_{3,\mathrm{thresh}}$. By multiplying $A_{3,\mathrm{thresh}}$ with the $E_r$ component in Eq. (1), we estimate that, at the threshold, the strength of the radial $E$ field within the nonlinear coating is

$$|\mathbf{E}^r_{\mathrm{thresh}}| = \sqrt{\frac{l_3(l_3+1)}{4\pi}} \frac{n^2 k_1 k_2 R^2}{|\Gamma^{l_3 m_3}_{l_1 m_1; l_2 m_2}||f_{l_1} f_{l_2}|} \frac{1}{\sqrt{Q_1 Q_2}} \frac{R}{\delta \chi^{(2)}_{rrr}}, \quad (5)$$

where $n$ is the refractive index of silica glass, and we have replaced $Y_{lm}(\theta, \varphi)$ in Eq. (1) with its average value $1/\sqrt{4\pi}$. To find a numerical estimate for $|\mathbf{E}^r_{\mathrm{thresh}}|$, we consider the three WG modes in Fig. 2(a) and use the exact microsphere and mode parameters given in the previous paragraph. We also make the following assumptions: (i) We conservatively assume that both $Q_1$ and $Q_2$ are $10^6$; (ii) from Fig. 2(a), we assume that $|f_{l1}|$ and $|f_{l2}|$ are 1; (iii) we assume that the nonlinear layer thickness is $\delta = 50$ nm and use $\chi^{(2)}_{rrr} = 14$ pm/V, the experimental result in Ref. [12]. Given these considerations, the threshold value for the pump radial electric field is estimated to be $1.8 \times 10^7$ V/m.

For the coupling configuration in Fig. 1(b), we can relate the threshold electric field strength to the pump power in the fiber taper. Since the electric field of a TM WG mode is dominated by its radial component, we can define an effective mode volume as $V_{\mathrm{eff}} = \iiint |E_r|^2 dv/[|E_r|^2_{\max}/2]$, where $|E_r|_{\max}$ is the maximum $|E_r|$ value. With this definition, we can estimate the pump energy stored in the microsphere as $\varepsilon_0 \varepsilon_r |\mathbf{E}|^2_{\mathrm{ave}} V_{\mathrm{eff}}/2$, where $|\mathbf{E}|_{\mathrm{ave}}$ is the average electric field strength. From coupled mode analysis [10,16], the pump power traveling in the fiber taper is related to the pump energy stored within the microsphere, which gives $P_{\mathrm{pump}} = \frac{\omega_3}{4Q_3} \varepsilon_0 \varepsilon_r |\mathbf{E}|^2_{\mathrm{ave}} V_{\mathrm{eff}}$. (Here we assume that the intrinsic cavity loss equals the coupling between the fiber taper and the microsphere [10].) To obtain a numerical value for $V_{\mathrm{eff}}$, we use Eq. (1) and $f_l(r)$ in Fig. 2 to calculate $E_r$, from which we determine that the effective volume for mode $|3\,248\,248\rangle$ is 190 $\mu\mathrm{m}^3$. Since the radial $E$ field is proportional to $f_l(r)$, from Fig. 2(a), we can roughly estimate $|\mathbf{E}|_{\mathrm{ave}}$ as 3 times the magnitude of $|\mathbf{E}^r_{\mathrm{thresh}}|$, which is the threshold value given by Eq. (5). By using $|\mathbf{E}^r_{\mathrm{thresh}}| = 1.8 \times 10^7$ V/m and assuming that $Q_3 = 10^6$, we find that it requires only a pump power of 9.7 mW to achieve parametric oscillation. If we increase all three $Q$ factors to $5 \times 10^6$, the threshold of parametric oscillation can be further reduced to only 78 $\mu$W. Such an ultralow threshold is not surprising, since pump threshold power scales as $1/Q_1 Q_2 Q_3$.

The proposed nonlinear microsphere can also lead to symmetry-enforced quantum entanglement. To illustrate this possibility, we focus on the degeneracy between the $|q, l, m\rangle$ and $|q, l, -m\rangle$ WG modes. This twofold degeneracy is due to time reversal symmetry and holds true even if the microsphere deviates from its spherical shape [17]. Now consider two high-$Q$ WG modes $|q_1, l_1, m_1\rangle$ at $\omega_1$ and $|q_2, l_2, 1 - m_1\rangle$ at $\omega_2$. The pump is a free space Gaussian beam at $\omega_3 = \omega_1 + \omega_2$ and is focused on the microsphere. As shown in Ref. [18], we can expand the Gaussian beam $|\Phi\rangle$ as a superposition of spherical waves, i.e., $|\Phi\rangle = \sum_{l_3=1}^{\infty} [\alpha_{l_3} |l_3, 1\rangle + \beta_{l_3} |l_3, -1\rangle]$, where the linear coefficients $\alpha_{l_3}$ and $\beta_{l_3}$ depend on the Gaussian beam parameters. (We drop the radial node number $q$, since the pump beam is no longer confined within the microsphere.) By following the analysis that leads to Eqs. (2) and (4), we observe that, as long as there exists one or more $l_3$ in the range of $|l_1 - l_2| \le l_3 \le l_1 + l_2$, the Gaussian beam, which contains a large number of $|l_3, 1\rangle$ components, can always generate signal and idler photons in the state of $|q_1, l_1, m_1\rangle |q_2, l_2, 1 - m_1\rangle$. Yet, due to the twofold degeneracy with respect to the signs of the $m$ quantum number, the $|l_3, -1\rangle$ pump component must also generate signal and idler photons in the state of $|q_1, l_1, -m_1\rangle |q_2, l_2, -1 + m_1\rangle$. The final state for the signal and idler photons is then a quantum entangled state of $A|q_1, l_1, m_1\rangle |q_2, l_2, 1 - m_1\rangle + B|q_1, l_1, -m_1\rangle |q_2, l_2, -1 + m_1\rangle$, where coefficients $A$ and $B$ depend on the parameters of the microsphere and the pump.

Finally, we discuss two practical yet important questions: the impact of the nonlinear film on the cavity $Q$

factors and the mechanism for tuning the on-resonance condition. For the nonlinear microsphere, the cavity loss comes from four sources [14]: WG mode radiation loss, bulk silica absorption, scattering due to the nonlinear film, and nonlinear molecule absorption. For silica microspheres with a radius above 10 $\mu$m, the WG mode radiation and the bulk silica absorption are small and can be ignored in our analysis [14]. To estimate the scattering loss due to the nonlinear film, we need two empirical parameters: the standard deviation $\sigma$ for film thickness and the correlation length $B$ for film roughness. From atomic force microscope images in Ref. [19], we can estimate that $\sigma = 2$ nm and $B = 25$ nm. By using Eq. (23) in Ref. [14], we find that the $Q$ factor due to scattering loss is typically $1 \times 10^7$. To account for absorption by the nonlinear molecules, we note that nonlinear polymer waveguide loss is typically 1 dB/cm [20]. By using this value and Eq. (24) in Ref. [14], we find that, for a 15 $\mu$m silica sphere coated with a 50 nm thick nonlinear film, the $Q$ factor due to nonlinear film absorption should be of the order of $2 \times 10^6$ or greater. In addition, Ref. [21] has reported polymer microresonators with $Q$ factors as high as $5 \times 10^6$. With these considerations, we conclude that it is realistic to expect nonlinear microspheres with $Q$ factors in the range of $10^6$.

Experimental work on silica microspheres has found that they can be slightly elliptical, with ellipticity in the range of 2% [8]. This slight change in shape has little impact on the $Q$ factors and the field distributions of the WG modes. It can, however, shift the WG mode frequency by an amount of $\Delta\omega/\omega = -e/6[1 - 3m^2/l(l+1)]$ [17], where $e$ is the ellipticity and $l$ and $m$ are the angular quantum numbers. For the set of WG modes shown in Fig. 2(a) and by assuming that $m = l$, this shape-induced frequency shift translates into a 24 GHz difference between $\omega_{3,0}$ and $\omega_{1,0} + \omega_{2,0}$, which, even though small, breaks the on-resonance condition. We need, therefore, a mechanism to adjust this frequency mismatch. We note that, by adjusting microsphere radius, the material dispersion of silica can produce a small relative shift between the resonant frequencies of the WG modes. In Fig. 2(b), we show the frequency detuning between the pump mode $|3\,248\,248\rangle$, signal mode $|1\,110\,110\rangle$, and idler mode $|2\,138\,138\rangle$ as a function of the microsphere radius. According to Fig. 2(b), we need only to adjust the microsphere radius by 60 nm to achieve a 24 GHz shift in frequency mismatch, and Ref. [22] has reported a subnanometer-level control over the microsphere radius through chemical etching. Other tuning techniques, such as changing the refractive index of the environment, may also be used.

In summary, we have demonstrated that a high-$Q$ nonlinear microsphere can achieve ultralow threshold parametric oscillation and symmetry-enforced quantum entanglement. It is also realistic to fabricate nonlinear microspheres with $Q$ factors in the range of $10^6$ and satisfy other constraints imposed by current technologies.

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