Ultralow Raman lasing threshold and enhanced gain of whispering gallery modes in silica microspheres

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Remarkable demonstrations of a cavity-induced giant reduction of the Raman lasing threshold in silica microspheres have been reported in Spillane *et al.* [Nature (London) **415**, 621 (2002)], yet a complete quantum electrodynamics (QED) treatment of this process including the rigorous expressions for the electromagnetic modes of the sphere is lacking. In this paper, we employ a cavity QED approach to describe Raman lasing in microscopic spherical dielectric resonators with third-order nonlinearity to derive explicit expressions for the Raman gain and lasing threshold in terms of mode-overlapping coefficients determined from full analytic expressions of the electromagnetic eigenmodes of a dielectric sphere. We present dependencies of the Raman lasing threshold on the the order n of overlapping whispering gallery modes for microspheres with diameter in the range 20–35 μm, demonstrating ultralow thresholds of between 70 and 90 μW, consistent with recent experimental results. We explain the reduction of the lasing threshold as due to an increase in the overlapping coefficients as the mode order is increased toward the regime where the modal energy is mostly confined to the surface region of the resonator. The presented theory can be easily generalized to any microcavity of regular morphology and for any combination of interacting whispering gallery modes.

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I. INTRODUCTION

Optical resonators and cavities with a very high quality factor (Q factor) have attracted significant interest as a testing ground for numerous fundamental quantum and nonlinear optical phenomena, as well as for a range of potential applications including single molecule detection and ultralow threshold laser cavities [1,2]. The ability of a resonator to trap photons for a duration spanning many optical cycles allows the study of coherent single atom-photon interactions, and can drastically modify the relaxation time of embedded atomic emitters. The dielectric spherical resonator is well known to have one of the highest Q factors out of any optical resonator due to the excitation of whispering gallery modes (WGM) [3] that are strongly confined near the resonator surface by total internal reflection [4].

One of the cavity-enhanced optical processes that has received significant attention is stimulated Raman scattering (SRS) and Raman lasing [4]. In SRS, a fraction of the energy of an optical pump is frequency shifted into a neighboring spectral region due to Raman scattering by atoms or molecules in the cavity. If the spectral region coincides with a resonant frequency of a cavity mode, the energy at the shifted frequency can occupy the mode (which is called a Stokes or anti-Stokes mode). Lasing occurs when the usual threshold condition is satisfied as the power of generation of the Raman shifted wave matches its losses from the cavity. Aside from a cavity-induced lowering of the Raman lasing threshold, of particular interest is that the shifted frequency may lie in a spectral range difficult to obtain in conventional semiconductor cavities through band-gap engineering. In this regard, dielectric microspheres stand to provide a new generation of highly compact and efficient laser sources in spectral regions usually difficult to access.

Numerous experimental demonstrations of the lowering of the SRS threshold in microresonators including silica microspheres [5], toroid microcavities [6,7], and spherical liquid microdroplets [8] have been attributed to cavity quantum electrodynamic (QED) effects [9,10]. These include a Purcell factor modification of the Raman transition rate of atoms in a cavity compared to in the bulk, and a Rabi frequency modulation of the Raman transition that governs the lasing threshold in cavities of very high Q factor [4].

Kippenberg *et al.* [6,7] have developed a semiclassical theory of SRS based upon coupled mode equations for the pump and Stokes electric field amplitudes. The intensity-dependent Raman gain is accounted for through an effective gain parameter that includes a mode-overlapping coefficient describing the spatial overlap of the pump and Stokes modes. Yet further physical insight is gained from a full quantum treatment where both the electromagnetic field and the molecular levels are quantized. For example, Wu *et al.* [4,11,12] developed a general QED theory of microcavity-enhanced Raman gain in the strong-coupling regime where the frequency linewidth of the Raman excitation is much larger than the linewidth of the cavity resonance. They considered only a single mode of the cavity to be occupied by a Stokes mode, and coupling between the Stokes mode and the pump is accounted for by an overlapping coefficient. In Ref. [4] they considered the specific case of Raman lasing of molecules in a Λ configuration in a spherical dielectric resonator with the pump and Stokes waves occupying separate modes of the cavity. They provide explicit expressions for the lasing threshold in terms of the cavity Q factor and cavity radius via a Rabi frequency that takes into account the finite linewidth of the Raman transition.

While these approaches have provided excellent physical insight into the Raman lasing process in microcavities, including quantitative dependencies of the lasing threshold on the cavity Q factor, Raman linewidth and pump-Stokes mode coupling coefficients, they do not explicitly take into account the full nature of the electromagnetic modes of the cavity or the nonlinear dielectric permittivity. In this regard, no insight is gained into how the lasing threshold depends on the order of interacting WGMs nor how to relate the lasing threshold to material parameters usually considered in optics. Therefore, in Ref. [13], a QED theory of Raman amplification in spherical dielectric cavities was developed starting from the interaction Hamiltonian explicit in the nonlinear dielectric permittivity (both second- and third-order nonlinearity were considered) and the electromagnetic modes of the sphere (from Mie theory [14]). While expressions were derived for the Raman lasing threshold in the strong-coupling regime in terms of the nonlinear dielectric permittivity and mode-overlapping coefficients, this study was limited to the overdamped regime, where the Rabi frequency of the Raman transition is much less than the rate of energy decay from the cavity; corresponding to Q factors in the range 10^4–10^7. Furthermore, in Ref. [13], calculations were specific to a second-order nonlinearity of the cavity. In the case of ultrahigh Q factor (10^8–10^{11}), the Rabi frequency is commensurate or much greater than the cavity decay rate, which is a likely condition in those experiments where ultralow Raman lasing threshold is observed (e.g., Spillane [2]).

In this paper we present a general QED theory of the SRS process in dielectric resonators in the strong-coupling and underdamped regime, where the Rabi frequency is commensurate or greater than the cavity decay rate. Raman scattering in the cavity is described as a two-photon process of interaction with three-level molecules of a Λ configuration, expressed through a third-order nonlinearity [15]. Our starting point is the associated Hamiltonian of third-order nonlinear interaction between the pump and Stokes modes. Explicit expressions are derived for the Raman lasing threshold and gain in terms of the cavity Q factor, decay rate, Raman linewidth, and the mode-overlapping coefficients. We provide calculations specific to spherical dielectric resonators which include evaluation of the rigorous expressions for the electromagnetic modes of the sphere. Specifically, the dependence of the lasing threshold on the mode order n is presented, which shows a decreasing lasing threshold as n is increased toward the regime where the pump and Stokes modes are mainly confined near the resonator surface. We predict ultralow thresholds of Raman lasing between 70 and 90 μW, consistent with previous experiments Ref. [5].

II. NONLINEAR WGM INTERACTION

We consider a cavity to be uniformly doped with three-level atoms of a Λ configuration that facilitate the interaction between separate whispering gallery modes (WGM) occupied by the pump and (anti-) Stokes fields [11,12], the latter corresponding to a (positive) negative Raman frequency shift of the pump. Occupation of a WGM by the pump is achieved through, say, fiber coupling with a resonant incident beam, while occupation of a separate WGM by the Stokes field requires that the Raman transition bring it into the frequency range of the corresponding cavity resonance. The Hamiltonian for the third-order nonlinear interaction between the WGMs in the cavity is given by Ref. [16]

$$H_{sp} = \chi^{(3)}_{sp}(\mathbf{E}_s \cdot \mathbf{E}_p)^2, \tag{1}$$

where $\chi^{(3)}_{sp}$ is the third-order nonlinear susceptibility that is uniform in the cavity, and we assume that the cavity is embedded in a uniform linear dielectric with $\chi^{(3)}_{sp} = 0$. $\mathbf{E}_s(\mathbf{r})$ and $\mathbf{E}_p(\mathbf{r})$ denote the electric fields of the pump and (anti-) Stokes modes. Hereafter the subscripts s and p refer to the (anti-) Stokes and pump modes, respectively, and we simply refer to Stokes modes without loss of generality.

The pump and Stokes modes at frequencies $\omega_{s,p}$ are quantized upon replacing the cavity eigenmodes $\mathbf{E}_{s,p}(\mathbf{r},t)$ by their corresponding quantum mechanical operators:

$$\mathbf{E}_{s,p}(\mathbf{r},t) = -i\left(\frac{\hbar\omega_{s,p}}{2\varepsilon_{s,p}}\right)^{1/2}\left[a^+_{s,p}(t) - a_{s,p}(t)\right]\mathbf{E}_{s,p}(\mathbf{r}), \tag{2}$$

where $a^+_{s,p}(a_{s,p})$ are the creation (annihilation) operators of photons occupying the s and p modes; $\mathbf{E}_{s,p}(\mathbf{r},t)$ are the normalized eigenfunctions of the spherical cavity [14] (see Appendix A). Substituting (2) into Eq. (1) we obtain the Hamiltonian for coupling between the pump and Stokes modes [11]:

$$H_{sp} = \hbar\{S_{sp}a_p a^+_s + \text{H.c.}\}. \tag{3}$$

Here H.c. denotes Hermitian conjugation, and

$$S_{sp} = t_s t_p(\omega_s\omega_p)^{1/2}\chi^{(3)}_{sp}B_c(\omega_s,\omega_p)(2)^{-1}(\varepsilon_s\varepsilon_p)^{-1/2} \tag{4}$$

is the integral coefficient of the Stokes-pump mode overlapping. $\varepsilon_{s,p}$ is the linear dielectric permittivity of the cavity (we may assume that $\varepsilon_s = \varepsilon_p$), $t_{s,p}$ are the amplitudes of the cavity modes obtained from the Mie theory [14], and $B_c(\omega_s,\omega_p)$ is the mode overlap integral taking the form [17],

$$B_c(\omega_s,\omega_p) = \frac{\int_V[\mathbf{E}^*_p(\mathbf{r})\cdot\mathbf{E}_s(\mathbf{r})]^2 d\mathbf{r}}{\int_V|\mathbf{E}_s(\mathbf{r})|^2 d\mathbf{r}\int_V|\mathbf{E}_p(\mathbf{r})|^2 d\mathbf{r}}, \tag{5}$$

where V is the cavity volume.

The electromagnetic energy loss from the cavity due to phonon-photon interactions is included in the Q factors (as a continuum model) by classical means [14,18].

The general photonic state in the cavity is $|\psi\rangle = \sum_{n_s,n_p} C_{n_s,n_p}|n_p,n_s\rangle$, where $|n_p\rangle$ ($|n_s\rangle$) represents the state corresponding to n_p (n_s) photons in the pump (Stokes) mode. We consider that the interaction occurs only in pairs consisting of one pump mode and one Stokes mode, and allow for the possibility that a number of Stokes modes may be excited due to the finite frequency width of the Raman transition Γ. Furthermore, we consider that only a single pump mode is excited by coupling with an incident laser beam of frequency ω_p. We seek the coefficients (C_{n_p}) and (C_{n_s}) of the eigenstates corresponding to a photon occupying the pump mode or a Stokes mode, respectively. Similarly to in Ref. [12] the Schrödinger equation with Hamiltonian Eq. (3) is integrated according to the Wigner-Weisskopf method [19] to obtain the set of linear differential equations:

$$\begin{aligned} i\hbar\frac{dC_p}{dt} &= \sum_{\{s\}} H^*_{sp}C_s\exp[i(\omega-\omega_s)t], \\ i\hbar\frac{dC_s}{dt} &= H_{sp}C_p\exp[-i(\omega-\omega_s)t], \end{aligned} \tag{6}$$

where the sum over $\{s\}$ is a summation over all of the excited Stokes modes. We consider now the limit that all of the excited

Stokes modes in the cavity except one contribute a negligible fraction of the total energy in the cavity (see Ref. [13]). Thus, retaining only a single Stokes mode s in the summation and integrating Eq. (6) we obtain

$$\frac{dC_p}{dt} = -\int_0^t \int_0^\infty \langle H_{sp} H_{sp}^* \rangle C_p(t') L(\omega_s - \omega, \gamma_s) \exp[-i(\omega_s - \omega)(t - t')] d\omega dt', \quad (7)$$

where $\langle H_{sp} H_{sp}^* \rangle / \hbar^2 = n_p(n_s + 1) S_{sp}^2$, γ_s is the Stokes mode linewidth in the cavity, $L(\omega_s - \omega, \gamma_s)$ is a Lorentzian centered at ω_s that describes the density of the Stokes mode in the frequency domain γ_s [19]. The initial condition is $C_p(t = 0) = 1$.

We consider the situation where $\Gamma > \gamma_s$ which holds for a cavity with high Q factor ($10^8 \sim 10^{11}$), that of $\Gamma < \gamma_s$ applying to the bulk or a low Q-factor cavity. We make the assumption that the mode order n of the WGM occupied by the pump and Stokes modes are large such that the electromagnetic energy is mainly concentrated at a thin surface layer of the microspheres (see below and Fig. 3; this regime corresponds to large WGM overlapping coefficients). Furthermore, the mode density of each WGM has been assumed to be described by a Lorentzian [19]. The probability of Raman events in the strong-coupling regime ($\Gamma \gg \gamma_s$) is obtained by integration of Eq. (7) over the cavity linewidth γ_s (Refs. [11,12]) to give $|C(t)|^2 \simeq \exp(-p_{s,1,2} t)$, where $p_{s,1}$ and $p_{s,2}$ are the roots of the following secular equation: $p_{s,1,2}(p_{s,1,2} - \gamma_s) + \beta_s = 0$ and $\beta_s = n_p(n_s + 1) S_{sp}^2$. When $\beta_s \gg (\gamma_s/2)^2$ we have $p_{s,1,2} = \gamma_s/2 \pm i\beta_s^{1/2}$, the imaginary part corresponding to oscillation of the rate of Raman events at the Rabi frequency [11,12]:

$$|p_{s,2}| = (\beta_s)^{1/2}. \quad (8)$$

When $(\gamma_s/2)^2 \gg \beta_s$, $p_{s,1}$ is real and the rate of Raman events is exponentially decaying at the inverse time constant $|p_{s,1}| = \beta_s/\gamma_s$. To realize Raman lasing in the cavity, the corresponding rate $p_{s,1,2}$ should be commensurate with the cavity decay time constant $\tau_R = \gamma_s^{-1}$, whereas for the bulk we take the decoherence time for the effective two-level Raman active molecule $\tau_R = \Gamma^{-1}$. Taking into account the definition of Q factor as the loading Q factor of the sphere coupled to the pump [3,18,20], the number of photons occupying the pump and Stokes mode is given by

$$n_{p,s} = I_{p,s} Q_{p,s} \sigma_{p,s} / (\hbar \omega_{p,s}^2) = P Q_{p,s} / (\hbar \omega_{p,s}^2), \quad (9)$$

where $I_{p,s}$ is the pump intensity in W/cm^2, P is the pump power in W, $\sigma_{p,s}$ are the extinction cross sections of the p,s eigenmodes [14,21], and ($Q_{p,s}$) the Q factors of the pump and Stokes modes, respectively.

Taking into account the linear dependence of the energy losses of each mode of the cavity for the whole Raman lasing decay rate, we have $\Gamma_c = \beta_s^{1/2} \gamma_c L(\omega_s - \omega_p, \gamma_c)$. Indeed, in the limit of high-order WGM, neighboring Stokes modes are almost completely frequency overlapping with mode s and may be excited by the pump. The factor $\gamma_c L(\omega_s - \omega_p, \gamma_c)$ where γ_c is the linewidth of the cavity, gives the approximate number of modes in the range $\omega_s - \omega_p$ that are excited. The total decay rate is then retrieved by multiplying the decay rate of a single Stokes mode s by the total number of WGMs excited by the Raman transition (where we have made the assumption that $\gamma_c \approx \gamma_s$). To obtain the dependence of decay rate on the pump intensity in the limit $n_s \gg 1$, we consider the conservation of the energy of photons: $\omega_p n_p + \omega_s n_s = \omega_p n_0$ where n_0 is the initial number of photons. The conservation condition for the number of photons is obtained from the Manley-Rowe equation, $\gamma_p n_p = \gamma_s n_s$. After some algebraic manipulation and in the limit of $\gamma_s < \gamma_p$, using Eq. (9), we can estimate the decay rate of SRS in the cavity:

$$\Gamma_c = \left(\frac{I_p Q_p \sigma_p}{\hbar \omega_p^2} \right) \frac{(\gamma_p \gamma_s)^{1/2}}{\left(1 + \frac{\omega_s \gamma_p}{\omega_p \gamma_s}\right)} S_{sp} L(\omega_s - \omega_p, \gamma_s). \quad (10)$$

Using Eq. (8), the threshold condition for Raman lasing in the cavity is [3,4,11]

$$(\beta_s)^{1/2} = \gamma_s. \quad (11)$$

The imaginary part of the decay rate describes the Raman lasing gain with the modulation frequency $\Omega_M = (\beta_s)^{1/2}$.

Taking into account Eqs. (9)–(11) for the threshold power of Raman lasing, we have

$$P_{th}^{st} = \frac{\hbar \omega_p^2}{Q_p} \left(\frac{\gamma_s}{\gamma_p} \right)^{1/2} \frac{\gamma_s}{S_{sp}} \left(1 + \frac{Q_s}{Q_p} \right). \quad (12)$$

III. CAVITY-ENHANCED GAIN OF WGM RAMAN LASING

The Raman gain in the cavity and bulk is $G_c = n_s^{th,c}/L_{\text{eff}}^c$ and $G_b = n_s^{th,b}/L_{\text{eff}}^b$, respectively, where $L_{\text{eff}}^{c,b}$ is the effective length of photon transport, and $n_s^{th,c}$ and $n_s^{\text{th},b}$ are the threshold photon numbers in the Stokes mode of the cavity c and bulk b, respectively. Raman lasing in the bulk must obey the threshold condition: $\Gamma = n_p^{\text{th},b}(n_s^{\text{th},b} + 1)\rho_0 V_b (S_{sp}^b)^2$, which characterizes the lifetime of photons in Stokes modes of the bulk, where ρ_0 is the bulk density of modes in the volume V_b [19]. S_{sp}^b denotes the perfect coupling in a large cavity or bulk and accounts for the optimal spatial overlapping between the input pump laser light and the Stokes mode. The integral coefficient of the mode overlapping is $B_{sp}^b = 1$ in a bulk medium. From Eqs. (8) and (11) we obtain the threshold condition which provides the relation between $n_s^{th,c}$ and $n_s^{th,b}$, thus the relative gain for Raman lasing is given by

$$\frac{G_c}{G_b} = \left(\frac{\Gamma}{\gamma_s} \right)^2 \frac{Q_s}{\omega_s \rho_0 V_b} \left(\frac{S_{sp}^c}{S_{sp}^b} \right)^2. \quad (13)$$

This expression describes the relative cavity enhancement of the Raman gain compared to the bulk. The relative gain in terms of the density of cavity modes relative to the bulk $D_c = L(\omega_s - \omega_p, \gamma_s)$ is [19]

$$\frac{G_c}{G_b} = \left(\frac{\Gamma}{\gamma_s} \right)^2 \frac{D_c}{\rho_0 V_b} [B_c(\omega_s, \omega_p)]^2. \quad (14)$$

This expression demonstrates that the enhancement is linear in D_c, and proportional to the square of the integral coefficients of spatial overlapping of the pump and Stokes modes $B_c(\omega_s, \omega_p)$ and the square of the frequency overlapping factor (modified Yokoyama-Brorson's factor [22]) Γ/γ_s. The concentration of

Raman active molecules N in the cavity can be obtained from $N = G_c\sigma_M^{-1}$, where σ_M is the molecular extinction cross section [14]. The extinction cross-section coefficients of the microcavity depend on both its material and morphological parameters. The enhancement factor is inversely proportional to the density of states of the Raman modes in the cavity ($\Gamma \gg \gamma_s$). For the photon spectrum of WGM Raman lasing see Appendix B.

IV. RESULTS AND DISCUSSION

Here we present calculations specific to dielectric spherical microspheres. To place our results in context with recent experimental work, we start by estimating the mode index n of WGM excited by the coupling fiber in Refs. [5,7] as given by $\rho = 2\pi r/\lambda_p \approx n$ [21] where r is the radius of the sphere, λ_p is the wavelength of the pump, and ρ is called the size parameter. Taking into account that the pump laser wavelength is $\lambda_p = 1.55\ \mu$m and $r \approx 20$–$35\ \mu$m we have that the range n of the WGM is approximately 84–147, and is the range of interest of n in our calculations. Further, the material parameters of the sphere are taken to be $\varepsilon_{s,p}^{1/2} = 1.44 - i10^{-7}$ and $\chi_{sp}^{(3)} = 1.4 \times 10^{-14}$ [23], and the pump of wavelength $\lambda_p = 1.55\ \mu$m is assumed to have a line width of 300 kHz. The laser pump central frequency is considered to be scanned repeatedly through a range of $\simeq$60 GHz around the resonance frequency of a single WGM with mode index n. Figure 1 shows the dependence of the power threshold of Raman lasing on the pump-Stokes mode interacting combination $TM_n - TM_{n-1}$ for radii of the sphere equal to (a) 20 μm and (b) 35 μm (Ref. [5]). The main interest in Fig. 1 is the extremely low threshold power for the interacting WGMs with mode numbers $80 \lesssim n \lesssim 160$. We see from Fig. 1 that for a microsphere radius of 20 μm that the threshold power of stimulated Raman lasing [Eq. (12)] decreases from 90 to 70 μW in the range $85 \leqslant n \leqslant 110$. For a microsphere radius of 35 μm (Figs. 1 and 2) the calculated threshold power P_{th}^{st} of Raman lasing for $125 \leqslant n \leqslant 160$ is $150 - 50\ \mu$W, while

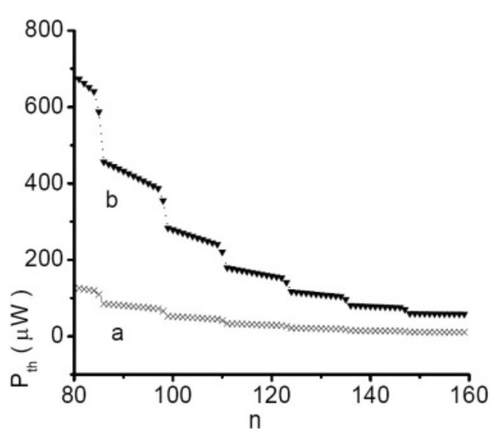

FIG. 1. Threshold power P_{th} of stimulated Raman lasing of overlapping modes $TM_n - TM_{n-1}$ with the radii of silica microsphere (a) 20 μm and (b) 35 μm. TM_n is the resonant pump mode. The refractive index of the silica microspheres is assumed $m = \varepsilon_s^{1/2} = 1.44 - i10^{-7}$ throughout.

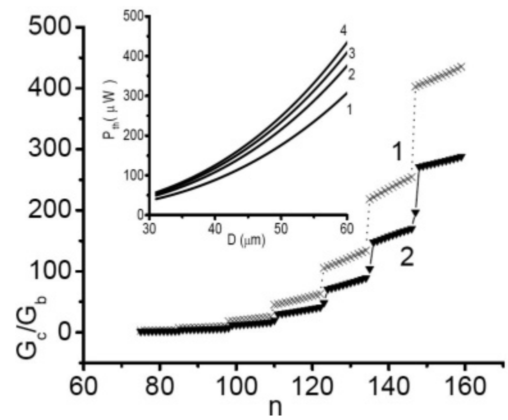

FIG. 2. G_c/G_b versus n for mode combinations: (1) $TM_n - TM_n^*$ and (2) $TM_{n-1} - TM_n$. The radius of the microsphere is $r = 35\ \mu$m. (Inset) Threshold of the stimulated Raman lasing versus diameter of the silica microsphere for WGM interaction combinations: (1) $TM_n - TM_n^*$, (2) $TM_{n-1} - TM_n$, (3) $TM_{n-2} - TM_n$, and (4) $TM_{n-3} - TM_n$ for $n = 140$.

that for $110 \leqslant n \leqslant 120$ is $\sim$150 μW, the former range of n giving good agreement with the measured threshold power in Ref. [5].

In Fig. 2 we present the Raman lasing gain in the cavity relative to in the bulk [Eq. (14)] as a function of the mode index number n for two different mode-overlapping combinations $TM_n - TM_n^*$ (where $*$ denotes nondegeneracy of the eigenmode [21]) and $TM_{n-1} - TM_n$ for a microsphere of radius $r = 35\ \mu$m. One can see that the relative gain grows within the mentioned range of n (Fig. 2) due to a dramatic increase of the mode-overlapping coefficient $B_c(\omega_s,\omega_p)$ (Fig. 3). It is remarkable that the relative gain G_c/C_b can exceed 10^2 and higher. In the limit of very large n, the Stokes mode and the pump mode become almost identical, ensuring the largest mode overlap. In the inset of Fig. 2 we present the dependence of lasing threshold on the diameter of the sphere for a number of different mode interaction combinations. This figure demonstrates a reduced lasing threshold as the interacting modes correspond to a more similar mode order n and an associated increased mode-overlapping coefficient. The reduction of the lasing threshold with a decreasing diameter of the sphere is easily explained due to an increase in Q factor of the pump and Stokes modes as the cavity size is reduced.

Another feature to be mentioned is that the threshold and gain curves are characterized by a steplike behavior. It is thought that these steplike features are related to the selection rules of interaction of the modes of the sphere (see, e.g., Ref. [13]) which separate regions of similar mode-overlapping coefficients. A detailed explanation of this phenomena is beyond the scope of the current paper, and will be the topic of a future work.

In practice, experimental measurement of the effective decay rate (i.e., cavity Raman linewidth) and cavity-enhanced gain of the Stokes modes Eq. (10) would provide the concentration of Raman active molecules as well as the energy-dependent Raman frequency shift of the ensemble of molecules in the microcavity.

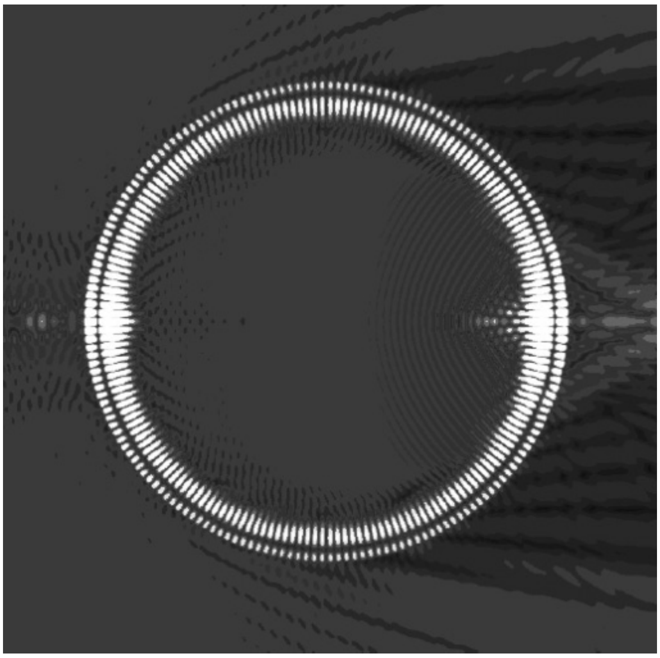

FIG. 3. The spatial distribution of electric field intensity of WGMs shows the mode overlapping, and the emission fields for a Raman microsphere laser (intrinsic pump quality factor of $Q = 10^8$, TM_{100}, $r = 20$ μm) excited near the threshold for stimulated Raman lasing.

V. CONCLUSION

In summary, on the basis of cavity QED theory we have investigated Raman lasing in high Q factor microcavities by coupling of quantized pump and (anti-) Stokes modes. We obtained analytical expressions for the normalized Raman lasing threshold and gain of a cavity which provides a good basis for experimental investigation of Raman lasing, Eqs. (10)–(14) being valid for a high Q cavity with third-order nonlinearity and having any regular shape. We have made calculations specific to dielectric microspheres which include the rigorous expressions for the electromagnetic modes (Mie theory), and have presented dependencies of the Raman lasing threshold and gain on the order and combination of interacting WGMs in spheres of various diameters. These results are in excellent agreement with the ultrahigh gain and ultralow threshold of Raman lasing in silica resonators observed in recent experiments [5].

The reduced lasing threshold occurs due to internal energy accumulation and nonlinear interaction between the cavity modes when both pump and (anti-) Stokes modes are resonant with WGMs. An increase in the order n of interacting WGMs is shown to reduce the lasing threshold due to a significant increase in the mode-overlapping coefficients. The threshold is further reduced as the diameter of the cavity is decreased due to an associated increase in the Q factor of the interacting pump and Stokes modes. Reducing the cavity size even further may improve the performance of these lasers. Nonlinear micro- and nanocavities, and hemispherical lenses are promising for these purposes [24].

Due to their small size and high Q factor, Raman microlasers based on WGM microspheres are attractive devices for potential applications including high-resolution spectroscopy, remote sensing, telecommunications, single-molecule sensors, and single-photon counters.

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APPENDIX A: EIGENMODES

In this section we provide exact expressions for WGM overlapping coefficients [Eq. (5)]. The TE and TM pump and Stokes modes in Eq. (5) are the solutions of the Helmholtz equation [14]:

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E}_j(\vec{r}) + k_j^2 \vec{E}_j(\vec{r}) = 0 \tag{A1}$$

for TE modes and

$$\vec{\nabla}^2 \vec{E}_j(\vec{r}) + k_j^2 \vec{E}_j(\vec{r}) = 0 \tag{A2}$$

for TM modes, respectively, obeying the boundary conditions and satisfying the normalization conditions within the cavity volume V [14]:

$$\int_V \vec{E}_i(\vec{r}) \vec{E}_j(\vec{r}) dV = \delta_{ij}. \tag{A3}$$

We assume that both the resonator and the surrounding medium are nonconductive and nonmagnetic. To calculate the integral coefficients of WGM overlapping the electric field in the cavity is written as a series of partial waves which are solutions of the Maxwell equations in spherical coordinates [14,21],

$$\mathbf{E}_{p,s} = E_0 \sum_{l=1}^{\infty} i^l \frac{2l+1}{l(l+1)} \left[t_l^{\mathrm{TE}} \mathbf{M}_{o1l}^3 - i t_l^{\mathrm{TM}} \mathbf{N}_{e1l}^3 \right], \tag{A4}$$

where E_0 is the amplitude of the incident laser beam that illuminates the cavity, i is the unit imaginary number, $\vec{r} = (r,\theta,\varphi)$ is the position vector in spherical coordinates, t_l^{TE} and t_l^{TM} are the partial wave amplitudes (Mie coefficients in the simplified case), and **M** and **N** are the eigenmodes of the cavity [partial waves, Eqs. (A5) and (A6)]. The functions in the series Eq. (A4) are given by

$$\mathbf{M}_{\sigma 1l}^{1,3}(\vec{r}) = \vec{\nabla} \times \vec{r} f_{\sigma ml}^{1,3}(\vec{r}), \tag{A5}$$

$$\mathbf{N}_{\sigma 1l}^{1,3}(\vec{r}) = \frac{1}{nk} \vec{\nabla} \times \vec{\nabla} \times \vec{r} f_{\sigma ml}^{1,3}(\vec{r}), \tag{A6}$$

where k is the free space wave number, $\vec{r}$ is the unit vector in the r direction, $\sigma =$ e or o identifying even or odd symmetry, respectively, and

$$f_{eml}^{f}(\vec{r}) = \psi_l(nkr) P_l^m(\cos\theta) \cos(m\varphi), \tag{A7}$$

where $\psi_l(nkr)$ are the Riccati-Bessel function of first order which should be replaced by the Riccati-Hankel function ζ_l^1 when the upper index of f is not 1 but 3, whereas the cosine function should be replaced by sine when $\sigma = o$. In Eq. (A7) the refractive index is n. The spherical eigenfunctions have the form,

$$\vec{H}_n = -im\sqrt{\frac{\epsilon_0}{\mu_0}}E_0\vec{N}^{1,3}_{\sigma mn} \quad \text{(A8)}$$

when

$$\vec{E}_n = E_0\vec{M}^{1,3}_{\sigma mn} \quad \text{(A9)}$$

and,

$$\vec{H}_n = -im\sqrt{\frac{\epsilon_0}{\mu_0}}E_0\vec{M}^{1,3}_{\sigma mn} \quad \text{(A10)}$$

when

$$\vec{E}_n = E_0\vec{N}^{1,3}_{\sigma mn}, \quad \text{(A11)}$$

which can be used to determine the magnetic fields. The continuity of the tangential components of both the electric and magnetic fields across the surface of the sphere provide us with the partial wave amplitudes [14].

We consider the electromagnetic eigenmode basis vectors according to Ref. [14] in the semiclassical QED theory of Raman scattering [17]. The main focus of our numerical calculations is the mode-overlapping coefficient as used in the theoretical investigation of Raman scattering in microcavities [17].

$$B_c(\omega_p,\omega_s) = \frac{\int_V |\mathbf{E}_p^*(\mathbf{r})|^2|\mathbf{E}_s(\mathbf{r})|^2 d\mathbf{r}}{\int_V |\mathbf{E}_s(\mathbf{r})|^2 d\mathbf{r} \int_V |\mathbf{E}_p(\mathbf{r})|^2 d\mathbf{r}}, \quad \text{(A12)}$$

where $\mathbf{E}_p$, $\mathbf{E}_s$, and ω_p and ω_s are the electric fields and eigenfrequencies of the pump and Stokes modes, respectively, and V is volume of the microscopic cavity. It can be seen from Eq. (A12) that the overlapping between the pump and Stokes modes is a function of $\mathbf{E}_p^*(\mathbf{r})\cdot\mathbf{E}_s(\mathbf{r})$, such that there is zero interaction if the modal fields are orthogonally polarized with respect to each other. Indeed, energy transfer between the pump and Raman photons is facilitated only by vibrations of the molecules of the medium, which is characterized by conservation of the number of photons [15,25] and there is no mechanism to facilitate energy transfer between the pump and Stokes photons of different polarizations when the medium is isotropic. We have assumed an isotropic medium in Eq. (5), and hence separated the $\mathbf{E}_p$ and $\mathbf{E}_s$ components in the same direction when calculating the matrix element for the interaction Hamiltonian between the pump and Stokes photons [11]. Indeed, when both $\mathbf{E}_p$ and $\mathbf{E}_s$ are plane waves, Eq. (5) is a simple and reasonable criteria for us to judge if Raman scattering is possible in an isotropic medium. The criteria of orthogonality of eigenfunctions is the Cauchy-Bunyakowski-Schwarz inequality [26]:

$$\frac{\left|\int_V \mathbf{E}_p^*(\mathbf{r})\cdot\mathbf{E}_s(\mathbf{r})d\mathbf{r}\right|^2}{\int_V |\mathbf{E}_p(\mathbf{r})|^2 d\mathbf{r}\int_V |\mathbf{E}_s(\mathbf{r})|^2 d\mathbf{r}} \leqslant 1. \quad \text{(A13)}$$

Equation (A13) shows the orthogonality between $\mathbf{E}_p$ and $\mathbf{E}_s$, which implies the cancellation among the components of scattering vector fields.

APPENDIX B: PHOTON SPECTRUM OF WGM RAMAN LASING IN STRONG-COUPLING REGIME

Since the decay rate of interacting Stokes and pump modes depends on the laser pump power, spatial mode overlapping has a Lorentzian form. We rewrite Eq. (7) to give an expression for the probability amplitude of the state with photon in mode $|s\rangle$ after the relaxation process. With the decay rate in the form of Eq. (10), we have for the intensity of spectra,

$$I(\omega) = \frac{\beta_s}{2\pi}\left(\frac{\gamma_s}{2}\right)^2\left[[\xi(\Delta-\xi)+\beta_s]^2+\left(\frac{\gamma_s}{2}\right)^2(\Delta-\xi)^2\right]^{-1}. \quad \text{(B1)}$$

In the tuned case when $\Delta = \omega - \omega_s = 0$ we have Eq. (B1), $\xi = \omega_p - \omega_s$. Then Eq. (B1) becomes

$$I(\omega_s) = \frac{1}{4}\frac{\gamma_s\beta_s^{st,sp}}{\xi}L\big(\beta_s^{sp,st}-\xi^2,\xi\gamma_s/2\big). \quad \text{(B2)}$$

For $\beta_s^{\mathrm{sp,st}}$ of spontaneous and stimulated Raman lasing we have

$$\beta_s^{\mathrm{sp}} = \frac{P_{\mathrm{th}}^{sp}Q_pS_{sp}^2}{\hbar\omega_p} \quad \text{(B3)}$$

and

$$\beta_s^{st} = \left(\frac{P_{\mathrm{th}}^{st}Q_p}{\hbar\omega_p}\right)^2\left(1+\frac{\omega_s\gamma_p}{\omega_p\gamma_s}\right)^{-2}\frac{\gamma_p}{\gamma_s}S_{sp}^2. \quad \text{(B4)}$$

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