Whispering-gallery-mode phase matching for surface second-order nonlinear optical processes in spherical microresonators

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The theory of surface-second-harmonic generation in a dielectric microsphere using whispering-gallery modes (WGMs) is developed. The second-order nonlinearity is restricted to the surface of the sphere. The coupling coefficients for a coupled-mode theory are derived and conditions for double resonance and phase matching are discussed for TE and TM polarizations. We demonstrate that phase matching of WGMs amounts to conservation of the angular momentum of the electromagnetic mode while at the same time we obtain an analytical expression for the coherence length.

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I. INTRODUCTION

The ability to manufacture spherical, toroidal, and microring cavities with ever increasing quality factors has motivated a large body of theoretical and experimental research in recent years. Currently, quality factors associated with optical whispering-gallery modes (WGMs) range from 10^7 to 10^{10} [1–3]. As a result, photons can experience the presence of atoms, particles at the rim of the cavity, or a nonlinear material response for a very long time, which opens new perspectives in quantum electrodynamics, sensing, and nonlinear optics [4–8]. Other current applications of WGMs are lasing [9], filtering [10], slow and fast light [11,12], and optomechanical effects [13–16]. For materials with bulk nonlinearities, Kerr bistability [17,18], frequency doubling [19,20], parametric oscillation [21], Raman effect [22], and nonlinear polarization conversion [23] have been demonstrated, and the optimal size of microspheres for linear and nonlinear interactions was investigated in Ref. [24]. A particularly important issue regarding microresonators is how to efficiently couple light into them—this was discussed in detail in Refs. [25–32]. A recent review on optical resonators with whispering-gallery modes can be found in Refs. [33,34].

Nonlinear optical studies with microspheres have also involved the scattering of plane waves, leading to third-harmonic [35] as well as second-harmonic generation for metallic [36] and dielectric spheres [37–44], even if the material is centrosymmetric. The latter is made possible by the breaking of symmetry at the surface of the sphere. While theoretical studies often focused on the small-sphere limit [37,39,45,46], the nonlinear scattering by spheres the size of several wavelengths was also treated in Refs. [47,48]. Similarly, the case of two-dimensional particles was analyzed in Refs. [49,50]. In nonlinear scattering experiments, phase-matching considerations are only important if many spheres are taken under consideration, as demonstrated theoretically and experimentally in Refs. [39,40,42,51,52].

The present paper addresses second-order nonlinear processes mediated by WGMs. As pointed out already, their high-quality factor can considerably enhance second-harmonic generation. Although this observation was made quite a few years ago, it was experimentally demonstrated only recently using microdisks of KTP [20]. Theoretically, the phenomenon was studied in the case where the nonlinearity extends throughout the bulk of the material filling the microdisk or microring resonator [19,53,54]. In this paper, however, we consider the effect of a nonlinearity on the surface of a centrosymmetric sphere, such as silica glass. Indeed, it is possible to coat such a sphere with a nonlinear material and one has the ability to choose the most efficient nonlinear molecule for a given range of frequencies. A longer-term goal could be to detect individual nonlinear molecules attached to the surface of the sphere. Electromagnetically, a single molecule manifests itself by a very localized nonlinear polarization. This possibility is encompassed by the theory presented here.

One of the major hurdles to overcome for an efficient interaction is to achieve phase matching between the two interacting waves while, ideally, having them both resonant with the microresonator [19,55]. In this regard, the frequency resonances of a dielectric sphere are well known, and useful asymptotic formulas are available for the spectrum of WGMs [56–58]. On the other hand, phase matching in microspheres has only been partially discussed. As we will establish here, phase-matching WGMs amounts to angular momentum conservation, in contrast to linear momentum conservation for plane waves. To our knowledge, this fundamental principle has not been enunciated before, probably because ring and disk geometries did not allow one to discern it fully. Of course, previous findings on phase matching are consistent with it [19,53,54,59].

The paper is organized as follows. In Sec. II, we introduce a coupled-mode description of the electromagnetic field when the dynamics can be reduced to a few WGMs near the fundamental and second-harmonic frequency. In this framework, the key ingredient to derive is the nonlinear coupling constant between WGMs. To this end, in Sec. III, we extend the classical treatment of spherical wave solutions of Max-

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well's equations to include the effect of a surface nonlinear polarization. This leads to an analytical expression for the coupling constants between fundamental and second-harmonic modes. Phase-matching conditions then arise from the requirement that the coupling be nonzero. In particular, we underline in Sec. IV the necessity to conserve angular, rather than linear, momentum. Furthermore, in the case of a nonuniformly distributed surface nonlinearity, we derive a simple analytical expression for the coherence length along the equator of the cavity. The explicit dependence of this coherence length on the sphere size is highlighted, and both phase-matching and quasi-phase-matching strategies are discussed. Finally, we conclude.

II. COUPLED-MODE DESCRIPTION

Let us consider a light wave at frequency ω being injected in a dielectric sphere with radius a via a tapered fiber. This mode of injection excites a WGM in the sphere. Through a surface second-order nonlinearity, one hopes to generate light at frequency 2ω. This requires the existence of another WGM near this frequency and to fulfill some phase-matching condition. Knowing the spherical modes $\mathbf{E}_\omega(\mathbf{r})$ and $\mathbf{E}_{2\omega}(\mathbf{r})$ that are most likely to participate to the dynamics, we can seek to express the field as

$$\mathbf{E} = \alpha_1(t)\mathbf{E}_\omega(\mathbf{r})e^{-i\omega t} + \alpha_2(t)\mathbf{E}_{2\omega}(\mathbf{r})e^{-2i\omega t} + \text{c.c.}, \tag{1}$$

and the problem is solved once the slow varying amplitudes $\alpha_i(t)$ are determined. A convenient way to normalize α_i is such that $|\alpha_i|^2$ is the radiated power. In this paper, we will derive equations of the form

$$\frac{d\alpha_1}{dt} + \Gamma_1\alpha_1 = i\kappa^*\alpha_1^*\alpha_2 e^{-i\Delta_{12}t}, \tag{2}$$

$$\frac{d\alpha_2}{dt} + \Gamma_2\alpha_2 = i\kappa\alpha_1^2 e^{i\Delta_{12}t}, \tag{3}$$

where $\Delta_{12}=\omega_2-2\omega_1$ is the detuning between the fundamental and second-harmonic WGM resonances, Γ_i are the width of these resonances, and κ is the nonlinear coupling between the two modes. The calculation of the coupling coefficient κ is the central result of the present work. Indeed, it gives the strength of the frequency conversion and we will see that it vanishes unless a precise phase-matching condition is satisfied.

III. SPHERICAL SOLUTIONS OF MAXWELL'S EQUATIONS AND COUPLED-MODE EQUATIONS

In this section, we extend the classical description of spherical wave solutions of Maxwell's equations to include the effect of a surface nonlinear polarization. With a dielectric sphere of radius a in a vacuum environment, Maxwell's equations can be written as

$$\nabla^2\mathbf{E} - \frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} = \mu_0\frac{\partial^2}{\partial t^2}[\mathbf{P} + \delta(r-a)\mathbf{P_{NL}}], \tag{4}$$

$$\nabla^2\mathbf{H} - \frac{1}{c^2}\frac{\partial^2\mathbf{H}}{\partial t^2} = -\nabla\times\frac{\partial}{\partial t}[\mathbf{P} + \delta(r-a)\mathbf{P_{NL}}], \tag{5}$$

$$\nabla\cdot\mathbf{E} = 0, \quad \nabla\cdot\mathbf{H} = 0, \tag{6}$$

where $\mathbf{P}$ is the linear polarization and $\mathbf{P_{NL}}$ is a surface nonlinearity. These vectorial equations can be turned into scalar ones thanks to the identity

$$\mathbf{r}\cdot(\nabla^2\mathbf{E}) = \nabla^2(\mathbf{r}\cdot\mathbf{E}) - 2\nabla\cdot\mathbf{E} = \nabla^2(\mathbf{r}\cdot\mathbf{E}).$$

This yields

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)(\mathbf{r}\cdot\mathbf{E}) = \mu_0\mathbf{r}\cdot\frac{\partial^2}{\partial t^2}[\mathbf{P} + \delta(r-a)\mathbf{P_{NL}}], \tag{7}$$

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)(\mathbf{r}\cdot\mathbf{H}) = -\mathbf{r}\cdot\left\{\nabla\times\frac{\partial}{\partial t}[\mathbf{P} + \delta(r-a)\mathbf{P_{NL}}]\right\}. \tag{8}$$

In the absence of $\mathbf{P_{NL}}$, both equations above have separable solutions of the form $Y_{lm}(\theta,\varphi)z_l(r)e^{-i\omega_{lp}t}$, where Y_{lm} is a spherical harmonic and z_l is a spherical Bessel function [60]. The indices l, m, and p are, respectively, the orbital, azimutal, and radial numbers. Furthermore, if $\mathbf{r}\cdot\mathbf{H}=0$, the solution is transverse magnetic and if $\mathbf{r}\cdot\mathbf{E}=0$, the solution is transverse electric.

Since $\mathbf{P_{NL}}$ is a small perturbation, we may seek a solution as a limited expansion of the form

$$\mathbf{r}\cdot\mathbf{E} = -\sum_{lmp}\sqrt{2Z_0 l(l+1)}\alpha_{lmp}^{(\mathrm{TM})}(t)z_l(r)Y_{lm}(\theta,\varphi)e^{-i\omega_{lp}t}, \tag{9}$$

$$\mathbf{r}\cdot H = \sum_{l'm'p'}\sqrt{\frac{2l'(l'+1)}{Z_0}}\alpha_{l'm'p'}^{(\mathrm{TE})}(t)z_{l'}(r)Y_{l'm'}(\theta,\varphi)e^{-i\omega_{l'p'}t}, \tag{10}$$

where $Z_0=\sqrt{\mu_0/\epsilon_0}$ is the vacuum impedance and the normalization is chosen such that $|\alpha_{lmp}|^2$ is the total power radiated by a given mode at infinity [60]. Furthermore, we assume that amplitudes depend slowly on time, in the sense that

$$\left|\frac{d\alpha_{lmp}}{dt}\right| \ll \omega_{lp}|\alpha_{lmp}|. \tag{11}$$

The form (2) and (3) of the equations for the α_{lmp} is easily anticipated. Our aim is to derive the coupling constants between the modes; these coupling constants characterize the strength of frequency conversion and also reflect phase-matching conditions. Throughout the remainder this paper, we will use primed indices to designate TE modes and unprimed indices for TM modes.

A. Transverse electric modes

Let us first consider a given transverse electric mode with quantum numbers L', M', and P' and denote its amplitude simply by α to avoid overloading the notation. Substituting Eq. (10) into Eq. (8), using Eq. (11) and the orthogonality of the spherical harmonics, we find

$$\frac{1}{r}\frac{d^2}{dr^2}(rz_{L'}) + \left[n^2k_{L'P'}^2 + \frac{2in^2k_{L'P'}}{c\alpha}\frac{d\alpha}{dt} - \frac{L'(L'+1)}{r^2}\right]z_{L'} = -\delta(r-a)\frac{J^{(\mathrm{TE})}}{\alpha}, \tag{12}$$

where $k_{L'P'}=\omega_{L'P'}/c$, $n=n(\omega_{L'P'})$ is the index of refraction resulting from the linear polarization $\mathbf{P}$, and

$$J^{(\mathrm{TE})} = \sqrt{\frac{Z_0}{2L'(L'+1)}}\int\int Y^*_{L'M'}\mathbf{r}\cdot\left(\boldsymbol{\nabla}\times\frac{\partial\mathbf{P}_{\mathbf{NL}}}{\partial t}\right) \times e^{i\omega_{L'P'}t}\sin\theta d\theta d\varphi. \tag{13}$$

We may simplify the left-hand side of the above equation by defining

$$K_{L'P'} \equiv k_{L'P'} + \frac{i}{c\alpha}\frac{d\alpha}{dt}. \tag{14}$$

Indeed, noting from Eq. (11) that $k_{L'P'}^2 + \frac{2ik_{L'P'}}{c\alpha}\frac{d\alpha}{dt} \approx K_{L'P'}^2$, Eq. (12) becomes

$$\frac{1}{r}\frac{d^2}{dr^2}(rz_{L'}) + \left[n^2K_{L'P'}^2 - \frac{L'(L'+1)}{r^2}\right]z_{L'} = -\delta(r-a)\frac{J^{(\mathrm{TE})}}{\alpha}. \tag{15}$$

For $r\neq a$, the solution of this equation is a combination of spherical Bessel functions, defined as

$$b_l(x) = \sqrt{\frac{2\pi}{x}}B_{l+1/2}(x), \tag{16}$$

where $B_l(x)$ is $J_l(x)$, $Y_l(x)$, $H_l^{(1)}(x)=J_l(x)+iY_l(x)$ or any other solution of Bessel's equation. Inside the sphere, to avoid divergence as $r\to 0$, the solution is of the form

$$z_{L'} = Aj_{L'}(nK_{L'P'}r), \tag{17}$$

while outside the sphere ($n=1$), the solution should be

$$z_{L'} = h_{L'}^{(1)}(K_{L'P'}r) \tag{18}$$

so as to ensure the correct asymptotic behavior $z_{L'}\propto e^{ikr}/r$ as $r\to\infty$. The constant A and $\frac{d\alpha}{dt}$ are determined through the continuity conditions at the boundary of the sphere $r=a$. On the one hand, the normal component of $\mathbf{H}$ is continuous, and hence

$$[z_{L'}]_-^+ = 0. \tag{19}$$

On the other hand, multiplying both sides of Eq. (15) by r, integrating from $r=a-h$ to $r+h$ and letting $h\to 0$, we get

$$\left[\frac{d}{dr}(rz_{L'})\right]_-^+ = a\left[\frac{dz_{L'}}{dr}\right]_-^+ + [z_{L'}]_-^+ = -a\frac{J^{(\mathrm{TE})}}{\alpha}. \tag{20}$$

Combining these two conditions, we find

$$\mathcal{F}_E(K_{L'P'}a) = \frac{J^{(\mathrm{TE})}}{K_{L'P'}\alpha}, \tag{21}$$

where (see Appendix A)

$$\mathcal{F}_E(K_{L'P'}a) \equiv nh_{L'}^{(1)}(K_{L'P'}a)\frac{j_{L'-1}(nK_{L'P'}a)}{j_{L'}(nK_{L'P'}a)} - h_{L'-1}^{(1)}(K_{L'P'}a). \tag{22}$$

In the following, it will be convenient to introduce the derivative of the function $\mathcal{F}_E$:

$$\mathcal{G}_E(x) \equiv \frac{d\mathcal{F}_E(x)}{dx}. \tag{23}$$

Note that Eq. (21) is just the characteristic equation for TE modes when $J^{(\mathrm{TE})}=0$. Hence, by definition of $k_{L'P'}$, we have

$$\mathcal{F}_E\left(k_{L'P'}a - i\frac{\Gamma_{L'P'}}{c}a\right) = 0, \tag{24}$$

where $\Gamma_{L'P'}$ is the decay rate of the mode. Since the quality factor is typically very large, we have $\Gamma_{L'P'}\ll k_{L'P'}c$ and a Taylor expansion of Eq. (24) to first order yields

$$\mathcal{F}_E(k_{L'P'}a) \approx i\frac{\Gamma_{L'P'}a}{c}\mathcal{G}_E(k_{L'P'}a). \tag{25}$$

Hence

$$\mathcal{F}_E(K_{L'P'}a) \approx \mathcal{F}_E(k_{L'P'}a) + \mathcal{G}_E(k_{L'P'}a)\frac{ia}{c\alpha}\frac{d\alpha}{dt} = \mathcal{G}_E(k_{L'P'}a)\frac{ia}{c}\left(\frac{1}{\alpha}\frac{d\alpha}{dt} + \Gamma_{L'P'}\right), \tag{26}$$

and Eq. (21) becomes

$$\frac{d\alpha}{dt} + \Gamma_{L'P'}\alpha \approx \frac{cJ^{(\mathrm{TE})}}{ik_{L'P'}a\mathcal{G}_E(k_{L'P'}a)}. \tag{27}$$

B. Transverse magnetic modes

Proceeding in the same way and with the same shorthand notation as in the previous section, we now consider the amplitude α of a transverse magnetic mode with quantum numbers L, M, and P. We now get

$$\frac{1}{r}\frac{d^2}{dr^2}(rz_L) + \left[n^2K_{LP}^2 - \frac{L(L+1)}{r^2}\right]z_L = \delta(r-a)\frac{J^{(\mathrm{TM})}}{\alpha}, \tag{28}$$

where

$$J^{(\mathrm{TM})} = \sqrt{\frac{1}{2Z_0L(L+1)}}\int\int \mu_0 Y^*_{LM}\mathbf{r}\cdot\frac{\partial^2\mathbf{P}_{\mathbf{NL}}}{\partial t^2}e^{i\omega_{LP}t}\sin\theta d\theta d\varphi \tag{29}$$

and where, as with the TE modes, we use

$$K_{LP} \equiv k_{LP} + \frac{i}{c\alpha}\frac{d\alpha}{dt}. \tag{30}$$

The solution is

$$z_L = \begin{cases} Aj_L(nK_{LP}r), & r < a, \\ h_L^{(1)}(K_{LP}r), & r > a. \end{cases} \tag{31}$$

From the continuity of the normal displacement field, and integrating Eq. (28) across $r=a$, we get

$$[n^2 z_L]_-^+ = 0, \quad \left[\frac{d}{dr}(rz_L)\right]_-^+ = a\frac{J^{(\mathrm{TM})}}{\alpha}. \tag{32}$$

This now yields

$$\mathcal{F}_M(K_{LP}a) = \frac{J^{(\mathrm{TM})}}{K_{LP}\alpha}, \tag{33}$$

where, this time,

$$\mathcal{F}_M(K_{LP}a) \equiv h_{L-1}^{(1)}(K_{LP}a) - h_L^{(1)}(K_{LP}a)\frac{j_{L-1}(K_{LP}a)}{nj_L(K_{LP}a)} - Lh_L^{(1)}(K_{LP}a)\left(1 - \frac{1}{n^2}\right). \tag{34}$$

We again denote the derivative of $\mathcal{F}_M$ by

$$\mathcal{G}_M(x) \equiv \frac{d\mathcal{F}_M(x)}{dx} \tag{35}$$

and, following the same reasoning as with the TE mode, we deduce

$$\frac{d\alpha}{dt} + \Gamma_{LP}\alpha \approx \frac{cJ^{(\mathrm{TM})}}{ik_{LP}a\mathcal{G}_M(k_{LP}a)}. \tag{36}$$

We thus see how, for both TE and TM modes, evolution equations for their amplitude can be derived. In order to make these equations more explicit and isolate the nonlinear coupling between the modes due to a second-order nonlinearity, we need to develop the expression for $J^{(\mathrm{TE})}$ and $J^{(\mathrm{TM})}$. This is what we do in the next section.

C. Nonlinear polarization produced by whispering-gallery modes

We now wish to derive explicit form for $J^{(\mathrm{TE})}$ and $J^{(\mathrm{TM})}$ appearing in Eqs. (27) and (36). To this end, let us consider a particular fundamental field composed of a transverse magnetic mode with quantum numbers l, m, and p, and a transverse electric mode with quantum numbers l', m', and p'. In order to avoid unnecessary indexes, we will write the amplitudes of the fundamental field as

$$\alpha_{l,m,p} \equiv \alpha_1 \quad \text{and} \quad \alpha_{l',m',p'} \equiv \alpha_{1'}. \tag{37}$$

We need only to focus on one arbitrary second-harmonic TM field with quantum numbers L, M, P and one second-harmonic TE field with quantum numbers L', M', and P'. We will thus write

$$\alpha_{L,M,P} \equiv \alpha_2 \quad \text{and} \quad \alpha_{L',M',P'} \equiv \alpha_{2'}. \tag{38}$$

The treatment of more complicated fields, involving more WGM's, can immediately be derived from the present case.

Mathematically, the defining feature of WGM's is that

$$l,l',m,m' \gg 1, \quad l-m, l'-m' = O(1).$$

In this case indeed, light is concentrated along the equator of the sphere. Exploiting this limit, we show in Appendix B that the fundamental field is given by

$$\mathbf{E} \sim \frac{-\sqrt{2Z_0}}{a}\left\{\alpha_1\left[lz_l\mathbf{e}_r + i\frac{d(rz_l)}{dr}\mathbf{e}_\varphi\right]Y_{lm}e^{-i\omega_{lp}t} + \alpha_{1'}k_{l'p'}az_{l'}Y_{l'm'}e^{-i\omega_{l'p'}t}\mathbf{e}_\theta\right\} \tag{39}$$

on the surface of the sphere $r=a$. Furthermore, assuming that both fundamental frequencies are close to some given pump frequency ω, we also have

$$|l-l'| \ll l.$$

We can summarize the conditions that apply to the fundamental field by saying that

$$m,m',l,l',n_1k_{l'p'}a \sim n_1ka + O((n_1ka)^{1/3}), \tag{40}$$

where $k \equiv \omega/c$ and $n_1 \equiv n(\omega)$, as follows from Eq. (B1). Obviously, the same can be said of the second-harmonic modes, but with the substitution $k \to 2k$, $n_1 \to n_2 \equiv n(2\omega)$.

On the surface of a centrosymmetric material, the only nonzero components of the nonlinear susceptibility are $\chi_{\perp\perp\perp}$, $\chi_{\perp\parallel\parallel}$, and $\chi_{\parallel\perp\parallel}$, where $\perp$ and $\parallel$ refer to directions that are perpendicular and parallel to the surface, respectively, as in Refs. [45] and [49]. The surface nonlinear polarization is, therefore,

$$\begin{aligned}\mathbf{P_{NL}} &= \epsilon_0\chi^{(2)}:\mathbf{EE} \\ &= \frac{2Z_0\epsilon_0}{a^2}\left(\left\{\chi_{\perp\perp\perp}\alpha_1^2l^2z_l^2Y_{lm}^2e^{-2i\omega_{lp}t} + \chi_{\perp\parallel\parallel}\left[-\alpha_1^2\left(\frac{d(rz_l)}{dr}\right)^2Y_{lm}^2e^{-2i\omega_{lp}t} + \alpha_{1'}^2k_{l'p'}^2a^2z_{l'}^2Y_{l'm'}^2e^{-2i\omega_{l'p'}t}\right]\right\}\mathbf{e}_r \right. \\ &\quad + \chi_{\parallel\perp\parallel}\left(2i\alpha_1^2lz_l\frac{d(rz_l)}{dr}Y_{lm}^2e^{-2i\omega_{lp}t}\mathbf{e}_\varphi \right. \\ &\quad \left.\left. + \alpha_1\alpha_{1'}k_{l'p'}alz_lz_{l'}Y_{lm}Y_{l'm'}e^{-i(\omega_{lp}+\omega_{l'p'})t}\mathbf{e}_\theta\right)\right).\end{aligned}$$

Using Eq. (40) this can be simplified as

$$\begin{aligned}\mathbf{P_{NL}} \sim 2\epsilon_0Z_0k^2\left(n_1^2\left\{\left[\chi_{\perp\perp\perp}z_l^2 - \chi_{\perp\parallel\parallel}l^{-2}\left(\frac{d(rz_l)}{dr}\right)^2\right]\mathbf{e}_r + 2i\chi_{\parallel\perp\parallel}l^{-1}z_l\frac{d(rz_l)}{dr}\mathbf{e}_\varphi\right\}Y_{lm}^2\alpha_1^2e^{-2i\omega_{lp}t} \right. \\ + \chi_{\perp\parallel\parallel}z_{l'}^2Y_{l'm'}^2\alpha_{1'}^2e^{-2i\omega_{l'p'}t}\mathbf{e}_r \\ \left. + n_1\chi_{\parallel\perp\parallel}z_lz_{l'}Y_{lm}Y_{l'm'}\alpha_1\alpha_{1'}e^{-i(\omega_{lp}+\omega_{l'p'})t}\mathbf{e}_\theta\right),\end{aligned} \tag{41}$$

which allows us to evaluate $J^{(\mathrm{TM})}$. On the other hand, we have

$$\mathbf{r}\cdot\left(\boldsymbol{\nabla}\times\frac{\partial\mathbf{P_{NL}}}{\partial t}\right)=\frac{1}{\sin\theta}\left[\frac{\partial}{\partial\theta}\left(\sin\theta\mathbf{e}_\varphi\cdot\frac{\partial\mathbf{P_{NL}}}{\partial t}\right)-\frac{\partial}{\partial\varphi}\mathbf{e}_\theta\cdot\frac{\partial\mathbf{P_{NL}}}{\partial t}\right].$$

This can be simplified by noting, first, that $\mathbf{P_{NL}}$ oscillates in times at very nearly twice the fundamental frequency ω; second, spherical harmonics for which Eq. (40) holds vary much more rapidly in the azimuthal direction than along the polar coordinate; third, under the same assumption, they only have appreciable magnitude close to $\theta=\pi/2$. Hence $\sin\theta\sim 1$ and

$$\mathbf{r}\cdot\left(\boldsymbol{\nabla}\times\frac{\partial\mathbf{P_{NL}}}{\partial t}\right)\sim 2i\omega\frac{\partial}{\partial\varphi}(\mathbf{e}_\theta\cdot\mathbf{P_{NL}}) \tag{42}$$

Moreover, invoking Eq. (40) again, $\frac{\partial}{\partial\varphi}Y_{lm}Y_{l'm'}=i(m+m')Y_{lm}Y_{l'm'}\sim 2in_1kaY_{lm}Y_{l'm'}$. Eventually, we thus get

$$\mathbf{r}\cdot\left(\boldsymbol{\nabla}\times\frac{\partial\mathbf{P_{NL}}}{\partial t}\right)\sim -8n_1^2k^4a\chi_{\parallel\perp\parallel}z_lz_{l'}Y_{lm}Y_{l'm'}\alpha_1\alpha_{1'}\times e^{-i(\omega_{lp}+\omega_{l'p'})t}. \tag{43}$$

Similarly, we find

$$\mu\mathbf{r}\cdot\frac{\partial^2\mathbf{P_{NL}}}{\partial t^2}\sim -8Z_0k^4a\left\{n_1^2\left[\chi_{\perp\perp\perp}z_l^2-\chi_{\perp\parallel\parallel}l^{-2}\left(\frac{d(rz_l)}{dr}\right)^2\right]Y_{lm}^2\alpha_1^2e^{-2i\omega_{lp}t}+\chi_{\perp\parallel\parallel}z_{l'}^2Y_{l'm'}^2\alpha_{1'}^2e^{-2i\omega_{l'p'}t}\right\}. \tag{44}$$

Let us now consider the effect of this nonlinear polarization on a transverse electric mode (quantum numbers L', M', P') whose frequency is close to the second harmonic $k_{L'P'}\approx 2k$. We obtain

$$J^{(\mathrm{TE})}\approx\frac{-2ka\mathcal{G}_E(2k_{L'P'}a)}{c}\kappa_{\mathrm{EM}}\alpha_1\alpha_{1'}e^{i(\omega_{L'P'}-\omega_{lp}-\omega_{l'p'})t},$$

where

$$\kappa_{\mathrm{EM}}=\frac{\sqrt{2/Z_0}n_1^2k^2z_lz_{l'}}{\epsilon_0n_2a\mathcal{G}_E(2k_{L'P'}a)}\int\int\chi_{\parallel\perp\parallel}Y_{L'M'}^*Y_{lm}Y_{l'm'}\sin\theta d\theta d\varphi, \tag{45}$$

and we recall that $n_2\equiv n(2\omega)$. On the other hand, for a transverse magnetic mode (L,M,P) we find

$$J^{(\mathrm{TM})}\approx\frac{-2ka\mathcal{G}_M(2k_{LP}a)}{c}[\kappa_{\mathrm{MM}}\alpha_1^2e^{i(\omega_{LP}-2\omega_{lp})t}+\kappa_{\mathrm{EE}}\alpha_{1'}^2e^{i(\omega_{LP}-2\omega_{l'p'})t}],$$

where

$$\kappa_{\mathrm{MM}}=\frac{\sqrt{2/Z_0}n_1^2k^2z_l^2}{\epsilon_0n_2a\mathcal{G}_M(2k_{LP}a)}\int\int\left[\chi_{\perp\perp\perp}-(lz_l)^{-2}\left(\frac{d(rz_l)}{dr}\right)^2\chi_{\perp\parallel\parallel}\right]\times Y_{LM}^*Y_{lm}^2\sin\theta d\theta d\varphi \tag{46}$$

and

$$\kappa_{\mathrm{EE}}=\frac{\sqrt{2/Z_0}k^2z_{l'}^2}{\epsilon_0n_2a\mathcal{G}_M(2k_{LP}a)}\int\int\chi_{\perp\parallel\parallel}Y_{LM}^*Y_{l'm'}^2\sin\theta d\theta d\varphi. \tag{47}$$

Expressions (45)–(47) constitute the main result of this article. Bearing in mind that $\chi_{\perp\perp\perp},\ldots$ are surface susceptibilities (unit: $\mathrm{m^2\,V^{-1}}$), the dimension of the coupling coefficients is $\mathrm{W^{-1/2}\,s^{-1}}$, consistently with the definition of the amplitudes α_i.

D. Coupled-mode equations

Introducing the detunings $\Delta_{\mathrm{EM}}=\omega_{L'P'}-\omega_{lp}-\omega_{l'p'}$, $\Delta_{\mathrm{MM}}=\omega_{LP}-2\omega_{lp}$, and $\Delta_{\mathrm{EE}}=\omega_{LP}-2\omega_{l'p'}$, we have thus derived

$$\left(\frac{d}{dt}+\Gamma_{L'P'}\right)\alpha_{2'}=i\kappa_{\mathrm{EM}}\alpha_1\alpha_{1'}e^{i\Delta_{\mathrm{EM}}t}, \tag{48}$$

$$\left(\frac{d}{dt}+\Gamma_{LP}\right)\alpha_2=i\kappa_{\mathrm{MM}}\alpha_1^2e^{i\Delta_{\mathrm{MM}}t}+i\kappa_{\mathrm{EE}}\alpha_{1'}^2e^{i\Delta_{\mathrm{EE}}t} \tag{49}$$

for the TE and TM second-harmonic amplitudes, respectively. Finally, if there were no losses, we would have

$$\frac{d}{dt}(|\alpha_1|^2+|\alpha_{1'}|^2+|\alpha_2|^2+|\alpha_{2'}|^2)=0, \tag{50}$$

and we can directly deduce from this that the evolution equations for the fundamental amplitudes α_1 and $\alpha_{1'}$ are

$$\left(\frac{d}{dt}+\Gamma_{l'p'}\right)\alpha_{1'}=\frac{i}{2}\kappa_{\mathrm{EM}}^*\alpha_{2'}\alpha_1^*e^{-i\Delta_{\mathrm{EM}}t}+i\kappa_{\mathrm{EE}}^*\alpha_2\alpha_{1'}^*e^{-i\Delta_{\mathrm{EE}}t}, \tag{51}$$

$$\left(\frac{d}{dt}+\Gamma_{lp}\right)\alpha_1=\frac{i}{2}\kappa_{\mathrm{EM}}^*\alpha_{2'}\alpha_{1'}^*e^{-i\Delta_{\mathrm{EM}}t}+i\kappa_{\mathrm{MM}}^*\alpha_2\alpha_1^*e^{-i\Delta_{\mathrm{MM}}t}. \tag{52}$$

The damping rates appearing in these coupled equations should include all losses suffered by the WGMs. So far, in the present theory, we have implicitly only been considering radiation losses. However, as the size of sphere increases, additional loss mechanisms become comparable or even exceed these radiation losses. These arise mainly from Rayleigh scattering by imperfections on the surface of the sphere and absorption losses [24,61]. One should therefore include these effect by letting

$$\Gamma_{lp}\rightarrow\Gamma_{lp}+\Gamma_{\mathrm{lp,Rayleigh}}+\Gamma_{lp,\mathrm{absorption}}$$

in Eq. (52) and similarly in Eqs. (48), (49), and (51). In Appendix C, we show how the theory can easily be modified to include absorption losses.

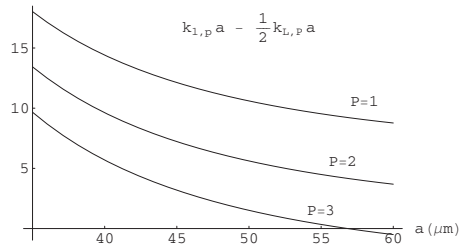

FIG. 1. Doubly resonant, phase-matched microsphere $(k_{l,p}-\frac{1}{2}k_{L,P})a$ as a function of sphere radius a for $l=650$, $p=1$, $L=2l$, and $P=1,2,3$. The choice $L=2l$ ensures phase matching for WGM's with $m=l$ and $M=L$. In order to achieve $k_{l,p}=\frac{1}{2}k_{L,P}$, a higher radial wave number must be assumed for the second harmonic. At double resonance ($a\approx 57$ μm) the fundamental wavelength is approximately 782 nm. See Appendix D for the Sellmeyer formula $n(\omega)$ used.

IV. PHASE MATCHING AND COHERENCE LENGTH

If the nonlinear susceptibility is uniform over the dielectric sphere, then from Eq. (45) we have

$$\kappa_{\mathrm{EM}} \propto \chi_{\parallel\perp\parallel}\int\int Y^{*}_{L'M'}Y_{lm}Y_{l'm'}\sin\theta d\theta d\varphi \tag{53}$$

and similarly for κ_{MM} and κ_{EE} from Eqs. (46) and (47), respectively. The integral above vanishes unless

$$M'=m+m', \quad |l-l'|\le L'\le l+l', \tag{54}$$

which are the selection rules for composing angular momenta in quantum mechanics. These are the phase-matching conditions for second-order nonlinear processes involving WGM's. Hence, in the present setting, phase matching amounts to conserve the angular momentum of the electromagnetic wave, in contrast to linear momentum for plane wave mixing.

In addition to Eq. (54), another condition for Eq. (53) not to vanish is that

$$L'+l+l'\in 2\mathbb{Z}. \tag{55}$$

This corresponds to the conservation of parity of the wave functions with respect to $\theta=\pi/2$.

Applying for example the selection rule above to a TM fundamental mode with $m=l$ and its TM second harmonic with $M=L$, matching requires $L=2l$. Such a choice corresponds to WGM's that are maximally concentrated on the equator and which are most naturally excited by a tapered fiber. Note that the phase-matching condition stated in Ref. [20] for toroidal resonators is consistent with Eq. (54) but only $m=l$ is achieved there, preventing conservation of angular momentum to be fully discerned. With the phase-matching condition $L=2l$, double resonance requires that $k_{2l,P}=2k_{l,p}$, where p and P are the radial numbers of the fundamental and second harmonic modes, respectively. In Fig. 1, we examine this possibility graphically for $p=1$ and various values of P. With the refraction index of silica, a fundamental wavelength around 800 μm and a sphere radius around 55 μm, we find that it is necessary to assume a radial number $P=3$ for the second harmonic. This is similar to Ref. [62]. Figure 1 also shows that phase matching and double resonance only occur simultaneously for specific sphere sizes only.

In a previous work [59], two counterpropagating fundamental WGMs (i.e., with $m_1=m$, $m_2=-m$) were found to be automatically phase matched with a second-harmonic $M=0$ mode. This obviously agrees with the condition of angular momentum conservation (54).

From the discussion above, it is clear that achieving simultaneously phase matching and double resonance is possible but probably rather difficult in practice. An alternative to phase matching is quasi-phase-matching. This can be achieved by covering the sphere only partly with a nonlinear material, or covering it with a periodic pattern of nonlinear material. In order to cover the sphere correctly, it is necessary to determine the coherence length. Focusing on the case $m=l$, $M=L,\ldots$, let us restrict the integration in Eqs. (45)–(47) to an angular sector of the sphere with arclength ℓ along the equator. We get

$$\kappa_{\mathrm{EM}} \propto \sin\frac{L'-l-l'}{2a}\ell, \quad \kappa_{\mathrm{MM}} \propto \sin\frac{L-2l}{2a}\ell,$$

$$\kappa_{\mathrm{EE}} \propto \sin\frac{L-2l'}{2a}\ell.$$

For κ_{MM}, for example, the coherence length is therefore

$$\ell_c=\frac{\pi a}{L-2l}. \tag{56}$$

As we move away from the equator, the coherence length is reduced by a factor $\sin\theta$.

It is possible to obtain an analytical approximate expression for ℓ_c. Given a fundamental frequency ω (wave number k), the orbital numbers l and L for the WGM's closest to the fundamental and second harmonics can be inferred from Eq. (B1). Asymptotically, we have

$$l\sim n_1 ka-\alpha_p(2n_1ka)^{1/3}+\epsilon_1, \tag{57}$$

$$L\sim 2n_2 ka-\alpha_p(4n_2ka)^{1/3}+\epsilon_2, \tag{58}$$

where ϵ_1, ϵ_2 ensure that l and L be integer. Hence,

$$\ell_c^{-1}=\frac{L-2l}{\pi a} \sim \frac{2(n_2-n_1)k}{\pi}\left[1+\frac{n_1^{1/3}-(n_2/4)^{1/3}}{n_2-n_1}\frac{2^{1/3}\alpha_p}{(ka)^{2/3}}\right]. \tag{59}$$

This formula is illustrated in Fig. 2, which demonstrates a strong dependence of the coherence length on the sphere size—and therefore not only with material dispersion. Note the difference with free space propagation. In the limit of a very large radius ℓ_c^{-1} tends to the bulk value $2(n_2-n_1)k/\pi$.

Phase matching or quasi-phase-matching implies double resonance for the two interacting waves. However, the two WGMs taking part in SHG are generally detuned by a finite amount Δ_{12} from 2:1 resonance. The appropriate values of l and L can be determined from the large-l asymptotic expansion of $k_{l,p}$ derived in Ref. [57] and which includes terms up

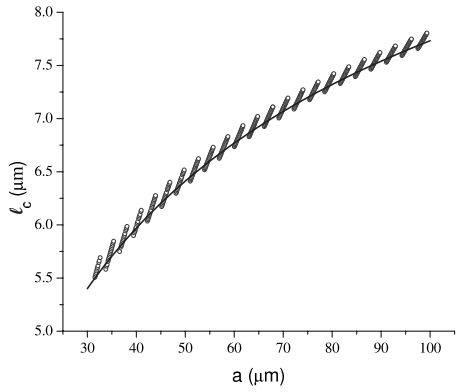

FIG. 2. Coherence length ℓ_c as a function of sphere radius. Continuous line: analytical formula (59); circles: numerically computed values from an eight-order asymptotic expansion relating mode frequency and mode orbital and radial numbers [57]. The fundamental wavelength is $\lambda=800$ nm. Only modes with $m=l$, $M=L$, and $p=P=1$ are considered.

to $O(l^{-8/3})$ inclusive [formula (B1) contains the first three terms of it]. To numerically compute ℓ_c as a function of the sphere radius a in Fig. 2, we proceeded as follows: We set the fundamental wavelength to 800 nm and chose a and l such that we got perfect resonance for that frequency. The values of L were then determined for these radii such that Δ_{12} was less than 10 MHz and the coherence length was deduced from Eq. (56). The jumps in ℓ_c as a function of a seen in Fig. 2 result from the discreteness of l and L. Such discrete character combined with the dependence of ℓ_c on the radius underscore the fundamental difference between the present situation and the nonlinear mixing of freely propagating plane waves.

Quasi-phase-matching would require to cover the sphere, for instance, with two types of domain slices, one with the nonlinear molecular dipole pointing outwards and the adjacent one with the nonlinear molecular dipole pointing inwards. Such periodic distribution of domains would lead to the largest conversion efficiency possible in such type of microresonators. Note that, from Eq. (56), in such a periodic configuration, the sphere perimeter is automatically equal to an even number of ℓ_c. Furthermore, quasi-phase-matching allows one to use the lowest order radial modes for both fundamental and second-harmonic frequency.

V. CONCLUSION AND OUTLOOK

In conclusion, we have developed a coupled mode equation theory to study second-order nonlinear generation of whispering-gallery modes when the nonlinear material is localized on the sphere surface. We have obtained explicit analytical expressions for the nonlinear coupling coefficients for the TE and TM polarizations. These expressions are used to establish the phase-matching condition as a conservation of the angular momentum of the electromagnetic wave. We showed that perfectly phase-matched, doubly resonant microresonators only assume some very specific radii. Furthermore, for typical experimental values—fundamental wavelength $\lambda\approx 800$ nm, $a\approx 50$ μm—phase matching is only possible between modes with different radial number.

Alternatively, we obtained the coherence length and showed that quasi-phase-matching is possible, the length of the equator being an even number of coherence lengths for WGM's. This coherent length depends significantly on the sphere size. Moreover, this dependence is discontinuous on account of the discreteness of the mode spectrum.

The theory of this paper treats the case of continuous waves. However, the coupled mode approach used could easily be extended to consider the case of a pulsed injection through the tapered fiber with a frequency bandwidth exceeding the free spectral range between consecutive WGMs. In this case, Eq. (1) in Sec. II must be extended to include enough fundamental modes and their second harmonics to cover the pulse bandwith.

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APPENDIX A: DERIVATION OF EQ. (21)

The continuity conditions (19) and (20) give

$$Aj_{L'}(nK_{L'P'}a) - h_{L'}^{(1)}(K_{L'P'}a) = 0, \tag{A1}$$

$$Anj'_{L'}(nK_{L'P'}a) - h_{L'}^{(1)\prime}(K_{L'P'}a) = \frac{J^{(\mathrm{TE})}}{K_{L'P'}\alpha}. \tag{A2}$$

Eliminating A from the latter, we get

$$nh_{L'}^{(1)}(K_{L'P'}a)\frac{j'_{L'}(nK_{L'P'}a)}{j_{L'}(nK_{L'P'}a)} - h_{L'}^{(1)\prime}(K_{L'P'}a) = \frac{J^{(\mathrm{TE})}}{K_{L'P'}\alpha}. \tag{A3}$$

Finally, using the identity

$$b'_l(x) = b_{l-1}(x) - \frac{l+1}{x}b_l(x), \tag{A4}$$

valid for all spherical bessel functions, we obtain Eq. (21).

APPENDIX B: ASYMPTOTICS EXPANSIONS FOR LARGE ORBITAL AND AZIMUTAL NUMBERS

In the large-l limit, the positions of the WGM resonances are given asymptotically by [58]

$$nk_{l,p}a = \nu + 2^{-1/3}\alpha_p\nu^{1/3} - \frac{B}{(n^2-1)^{1/2}} + O(\nu^{-1/3}), \tag{B1}$$

where $\nu=l+1/2$, α_p is the pth root of the Airy function $\mathrm{Ai}(-z)$, and

$$B = \begin{cases} n & \text{for TE modes,} \\ 1/n & \text{for TM modes.} \end{cases}$$

In Ref. [57], the coefficients of the expansion above were computed up to $O(\nu^{-8/3})$ and in Ref. [56], the width and

strength of these resonances is also given up to $O(\nu^{-2/3})$. Furthermore, we have [63]

$$J_\nu(ka) + iY_\nu(ka) \sim \frac{e^{-\nu(\beta-\tanh\beta)}}{\sqrt{2\pi\nu\tanh\beta}} - i\frac{e^{\nu(\beta-\tanh\beta)}}{\sqrt{\frac{1}{2}\pi\nu\tanh\beta}},$$

where $\cosh\beta=\frac{\nu}{ka}$ or

$$\beta \sim \operatorname{arccosh}(n) - \frac{2^{1/3}n\alpha_p}{\sqrt{n^2-1}}\nu^{-2/3} + \frac{nB}{n^2-1}\nu^{-1} + O(\nu^{-4/3}).$$

Hence, $Y_\nu(ka) \gg J_\nu(ka)$ and

$$h_l^{(1)}(ka) \sim -2i\cosh\beta\frac{e^{\nu(\beta-\tanh\beta)}}{\nu\sqrt{\sinh\beta}} \sim \frac{-2ine^{\nu(\beta-\tanh\beta)}}{(n^2-1)^{1/4}\nu}.$$

Note that, on account of Eq. (B1), we may write $\cosh\beta=n+O(\nu^{-1/3})$ and $\sinh\beta=\sqrt{n^2-1}+O(\nu^{-1/3})$, but the argument of the exponential above, being multiplied by ν, should be expressed with $O(\nu^{-1})$ accuracy for a precise evaluation.

Another useful formula was derived in Refs. [31,64] concerning spherical harmonics Y_{lm} whith $l,m\gg1$ and $q\equiv l-m=O(1)$. In this limit, the magnitude of Y_{lm} is expected to be significant only in the neighborhood of $\theta=\pi/2$ and to decay rapidly away from it. This motivates the introduction of a new polar variable $s=l^{1/2}(\pi/2-\theta)$ and leads to

$$Y_{lm} = P_l^m(\theta)e^{im\varphi} \sim (-)^{l+q}\frac{2^{1/4}l^{1/4}}{(2\pi)^{3/4}}\frac{H_q(s)e^{-s^2/2}}{(2^q q!)^{1/2}}e^{im\varphi}, \quad \text{(B2)}$$

where H_q is Hermite's polynomial. The $(-)^{l+q}$ factor (absent in Ref. [31]) can be deduced by comparing $P_l^m(0)$ and $\frac{dP_l^m}{ds}(0)$ with the explicit representation of $H_q(s)$ (respectively, p. 334 and p. 775 in Ref. [63]).

We now turn to the large-l approximation for vector spherical harmonics. These are defined as [60]

$$\mathbf{X}_{lm} = \frac{1}{\sqrt{l(l+1)}}\mathbf{L}Y_{lm} \sim \frac{1}{l}\mathbf{L}Y_{lm},$$

where $\mathbf{L}$ is the vector angular momentum operator, defined by

$$\mathbf{L} \equiv i\left(\mathbf{e}_\theta\frac{1}{\sin\theta}\frac{\partial}{\partial\varphi} - \mathbf{e}_\varphi\frac{\partial}{\partial\theta}\right) \sim -l\mathbf{e}_\theta + il^{1/2}\mathbf{e}_\varphi\frac{\partial}{\partial s}.$$

Transverse magnetic mode are thus given by

$$\mathbf{H}^{(\mathrm{TM})} = \sqrt{2/Z_0}k\alpha^{(\mathrm{TM})}z_l\mathbf{X}_{lm}e^{-i\omega_{lp}t} \sim -\sqrt{2/Z_0}k\alpha^{(\mathrm{TM})}z_l Y_{lm}e^{-i\omega_{lp}t}\mathbf{e}_\theta \quad \text{(B3)}$$

and

$$\mathbf{E}^{(\mathrm{TM})} = \frac{iZ_0}{k}\nabla\times\mathbf{H}^{(\mathrm{TM})} \sim -\frac{\sqrt{2Z_0}}{r}\alpha^{(\mathrm{TM})}\left[lz_l\mathbf{e}_r + i\mathbf{e}_\varphi\frac{\partial(rz_l)}{\partial r}\right]Y_{lm}e^{-i\omega_{lp}t}. \quad \text{(B4)}$$

At the surface of the sphere,

$$\frac{\partial}{\partial r}(rz_l) = kah_{l-1}^{(1)}(ka) - lh_l^{(1)}(ka).$$

Hence, since $ka\sim l/n$, the radial and azimutal electric field components are of comparable size. Further away from the center of sphere, when $kr\gg l$, $\mathbf{E}^{(\mathrm{TM})}$ becomes more and more parallel to $\mathbf{e}_\varphi$, making the Pointing vector parallel to $\mathbf{e}_r$. On the other hand, the transverse electric mode $(\mathbf{E}^{(\mathrm{TE})},\mathbf{H}^{(\mathrm{TE})})$ is obtained by the transformation $(\mathbf{E}^{(\mathrm{TM})},\mathbf{H}^{(\mathrm{TM})}) \to (-Z_0\mathbf{H}^{(\mathrm{TE})},Z_0^{-1}\mathbf{E}^{(\mathrm{TE})})$ in Eqs. (B3) and (B4).

APPENDIX C: TREATMENT OF ABSORPTION LOSSES

In the presence of absorption losses, the index of refraction becomes complex and we can generally assume that, at a given frequency $n(\omega)=n_r+in_i$, with $n_i\ll n_r$. In Eq. (12), for instance, we now have

$$\begin{aligned} n^2k_{L'P'}^2 + \frac{2in^2k_{L'P'}}{c\alpha} &= n^2\left(k_{L'P'}^2 + \frac{2ik_{L'P'}}{c\alpha}\frac{d\alpha}{dt}\right) \\ &\approx n_r^2\left(1+\frac{2in_i}{n_r}\right)\left(k_{L'P'}^2 + \frac{2ik_{L'P'}}{c\alpha}\frac{d\alpha}{dt}\right) \\ &\approx n_r^2\left(k_{L'P'}^2 + \frac{2ik_{L'P'}}{c\alpha}\frac{d\alpha}{dt} + \frac{2ik_{L'P'}^2n_i}{n_r}\right) \\ &= n_r^2\left[k_{L'P'}^2 + \frac{2ik_{L'P'}}{c}\left(\frac{1}{\alpha}\frac{d\alpha}{dt} + \frac{n_i}{n_r}ck_{L'P'}\right)\right]. \end{aligned} \quad \text{(C1)}$$

Hence, after defining

$$\Gamma_{L'P',\mathrm{absorption}} \equiv \frac{n_i}{n_r}ck_{L'P'},$$

the analysis follows exactly the same steps as in Sec. III A, but with $\frac{1}{\alpha}\frac{d\alpha}{dt}$ replaced everywhere by $\frac{1}{\alpha}\frac{d\alpha}{dt}+\Gamma_{L'P',\mathrm{absorption}}$.

APPENDIX D: SELLMEYER FORMULA

In our calculations, we used

$$n^2(\lambda) = 1 + \sum_{i=1}^{3}\frac{B_i\lambda^2}{\lambda^2-L_i^2},$$

with

$$B_1 = 0.6961663, \qquad L_1 = 0.0684043 \ \mu\text{m},$$

$$B_2 = 0.4079426, \qquad L_2 = 0.1162414 \ \mu\text{m},$$

$$B_3 = 0.8974794, \qquad L_3 = 9.896161 \ \mu\text{m}.$$

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