Existence of a threshold for second-harmonic generation inside high-confinement microresonators as a consequence of the generalized creation and annihilation operators

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This work presents a quantum theory of the nonlinear optical process of second-harmonic generation (SHG) in one-dimensional microresonators. More specifically, we show how the manipulation of vacuum field fluctuations in high-confinement systems, leading to a spectrally (and spatially) modulated commutation relation for the photon's generalized formulations of their creation and annihilation operators, deeply affects SHG behavior and gives rise to a threshold level. The two main effects the modulated commutator has on this optical process are an inhibition of the SHG process at low pumping level and a significant (cubic) amplification of the second-harmonic signal production rate once the threshold is overcome (finally reaching the usual quadratic dependence at sufficiently high pumping level). Our predictions, which represent a concrete picture of a fractional quantum system, could be used to probe vacuum field fluctuations present in high-confinement microresonators and emphasize the fundamental importance of vacuum field fluctuations.

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I. INTRODUCTION

As we are now capable of miniaturizing electromagnetic (EM) and photonic systems to reach the nanometer scale [1–12], the reduction and control of quantum noise have become increasingly crucial. The ability to measure and control electromagnetic quantum noise in confinement structures is of great importance both for fundamental interests and for applications. Despite efforts to reduce thermal and EM noise through various fabrication techniques and measurement structures, there will always remain a minimal level of quantum noise in the form of vacuum field fluctuations in any system [13–15].

The existence of virtual photons generating quantum vacuum field fluctuations is predicted by Heisenberg's uncertainty principle. Even though their role has been qualitatively recognized [16–21], their spectral and spatial density, which is linked to their rate of creation and amplified within high-confinement systems due to resonance, have not been explicitly formulated. Instead, the theoretical approach is based on Purcell's and Kleppner's conjectures [17,22]; the former consists of replacing the free spectral range by the "linewidth," and the latter consists of replacing the electronic density of states by an *ad hoc* "photonic density of states." While founded on the concept of the density of states, these conjectures are, nevertheless, questionable in the case of resonators (open cavities) [23]. The main concern is that they distort the original definitions of modes. As pointed out in Sec. II, this might lead to confusing quasimodes (resonant modes), especially quasinormal modes, with eigenmodes. Moreover, these conjectures assume the possibility of continuously modulating (here, spectrally) the density of modes (states) in between resonant frequencies and, accordingly, in the vicinity of a single quasimode ("piling up" of modes near resonances) [23].

However, a quantized fluctuational electrodynamics formalism was recently developed to show how the density of states concept can be used to describe the electromagnetic field ladder operators so that they no longer exhibit the anomalies reported for resonant structures [24]. As an alternative and in contrast to Langevin's formalism to describe open cavities [25–29], our approach is based on the so-called modes of the universe [30–33]. This *ab initio* approach turns out to be markedly distinct from previous approaches [34,35]. Formally, this approach leads to the introduction of a modulated commutator [36] between the creation and annihilation operators for photons inside a microcavity and results in the generalized creation and annihilation operators [23,37]. As detailed in Sec. V, the application of this formalism turns out to be very simple and might be a very useful alternative to investigate various dissipative (open) systems.

In this work, we focus on the nonlinear optical process of second-harmonic generation (SHG). By appropriately pumping a microcavity containing a second-order nonlinear material, we can probe vacuum field fluctuations by studying how they affect both the signal and noise of the SHG output.

This paper is organized as follows. In Sec. II, the one-dimensional high-confinement open microcavity system is defined, and the physical significance of the modulated commutator is underlined. Next, in Sec. III, using the cavity's quasinormal modes to describe the fields, the relevant interaction Hamiltonian for SHG is obtained. In Sec. IV, Heisenberg's equation of motion is used to predict the temporal evolution of the creation and annihilation operators for SHG within the microcavity. Finally, in Sec. V, the results are summarized and discussed.

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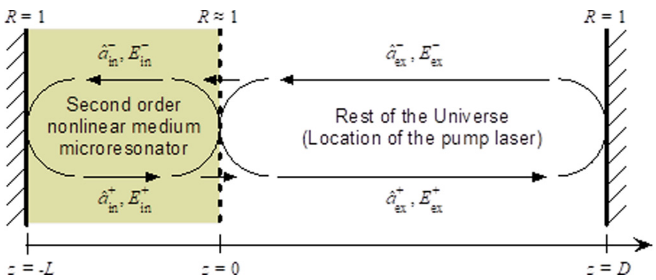

FIG. 1. Illustration of a one-dimensional universe conceived as a lossless closed cavity, bounded by perfectly conducting walls ($R = 1$ at $z = -L$ and $z = D$) including a second-order nonlinear resonator. The pump wave originates from the rest of the universe, between $z = 0$ and $z = D$. In the event of a high-finesse resonator, the semireflecting mirror at $z = 0$ is assumed to have a reflectance close to unity. The operators $\hat{a}$ represent the annihilation operators for each electric field component E. The subscripts "in" and "ex" refer to the resonator and the rest of the universe, respectively, while the superscripts "-" and "+" refer to the propagation direction.

II. PHYSICAL SIGNIFICANCE OF THE MODULATED COMMUTATOR IN MICROCAVITY SYSTEMS

As shown in Fig. 1, we consider a one-dimensional system at a temperature of 0 K enclosed between two perfect parallel reflectors with a reflectance of 1 at all frequencies. With the z axis defined as the direction normal to these surfaces, the reflectors are placed at $z = -L$ and $z = D$. An interface is located at $z = 0$ with a reflectance R almost equal to 1. This interface separates the microcavity of length L from the rest of the system. A nonlinear material is placed in the high-confinement region, and the pump wave originates from the region of length D. It is assumed that the entire system is composed of lossless materials.

It is important to underline that our formalism is based on the modes-of-the-universe point of view [30–33]. As shown in Fig. 1, in this approach the so-called universe is constituted by a single open planar microresonator, here incorporating a second-order nonlinear medium, and the "rest of the universe." In the event that $D \to \infty$, the microcavity has a negligible effect, and the rest of the universe is viewed as an immutable reservoir, which behaves as a canonical closed cavity where we assume that canonical commutation relations apply. This postulate about the reservoir is essential. Only the universe is a closed cavity. As such, true eigenmodes can be strictly defined only for the universe.

Such eigenmodes, having well-defined angular frequency ω, do not have any spectral width ("zero measure"). In the limit $D \to \infty$, the Fock states become very close together, and the ω values can be assumed to form (or be approximated by) a continuous set, so that ω can take practically any value. On the other hand, since the microcavity is open, the modes of the universe naturally extend spatially into the microcavity. At some specific angular frequencies ω_ν ($\nu = 1, 2, 3, \dots$), which ensure that an eigenmode reproduces itself (returns in phase) inside the microresonator after a single round trip, "quasimodes" can be formed in the vicinity of ω_ν. When $D \to \infty$, a large ("infinite") number of the universe's eigenmodes, having an angular frequency close to ω_ν, occur. Then, due to self-destructive interference, a quasimode is established, which is a superposition of an infinite number of eigenmodes (Ref. [38] showed that the weighting distribution is $\Lambda(\omega)$, given by Eq. (4)). Incidentally, the higher the finesse (or the quality factor Q) of the microresonator is, the closer the quasimodes are to a subset of the modes of the universe. It is noteworthy that each eigenmode contributes at least partially to the total quantum noise of the quasimode. As underlined in Sec. 3.3 of Ref. [39], in free space we can work with discrete sums over modes and replace the summations by suitable integrations when the cavity is sufficiently large so that there is practically a continuum of modes [40]. This approach is possible whenever there is an approximately continuous distribution of modes of interest. In summary, our formalism is equivalent to assuming that D can be as large as required but never model an unbounded universe. Here, $D \to \infty$ *a posteriori*. From this point of view, one can always implicitly keep the discrete picture of eigenstates and thus avoid the problem of the zero-measure bandwidth of eigenstates encountered in the truly continuous Fock-space mode description of an unbounded universe (see Sec. 10.10 in Ref. [34]). Incidentally, since the wave vectors of the continuous Fock space defined in Ref. [34] are here coincident, the definitions of creation and annihilation operators in that book do not apply properly to our point of view.

When the microcavity has a high quality factor, we can approximate that the quasinormal modes are identical to those found in a perfectly closed cavity of length L. The condition for resonant angular frequencies ω in the microcavity will be $L\omega = m\pi c$, with m being an integer and c being the speed of light in the nonlinear medium. The pump region has a length D chosen so that mode coupling to the microcavity is possible when D is several orders of magnitude larger than L. The pump region then acts as a reservoir and does not contain any confinement effects comparable to those of the microcavity. Experimentally, this condition is automatically met, as L is in the nanometer- or micrometer-scale.

Following Ueda and Imoto [36], we now define the four relevant operators to describe the pump photons as they travel through the system shown in Fig. 1 at normal incidence to the interface at $z = 0$. Those originating from the pump region traveling towards the microcavity (left) are represented by $\hat{a}^-_{\mathrm{ex}}$. The output traveling away from the microcavity (right) is represented by $\hat{a}^+_{\mathrm{ex}}$. We use $\hat{a}^-_{\mathrm{in}}$ and $\hat{a}^+_{\mathrm{in}}$ for photons within the microcavity, propagating to the left and to the right, respectively.

Because the pump region is empty and very large compared to the microcavity, the implicit postulate made by Ueda and Imoto, where the rest of the universe is considered to be an unbounded immutable reservoir and ω is continuous, appears to be suitable. This allows giving the commutator between the annihilation and creation operators for this region its canonical value [36],

$$\left[\hat{a}^-_{\mathrm{ex},\omega}, \hat{a}^{-\dagger}_{\mathrm{ex},\omega'}\right] = \left[\hat{a}^+_{\mathrm{ex},\omega}, \hat{a}^{+\dagger}_{\mathrm{ex},\omega'}\right] = \delta(\omega - \omega')\hat{I}. \tag{1}$$

However, recalling our point of view of a bounded universe and its discrete picture of eigenstates that resort to a quasicontinuous basis, Eq. (1) is regarded here as a suitable approximation (see Eqs. (41) and (42) and the text that follows in [23]). Nevertheless, this assumption naturally leads to the

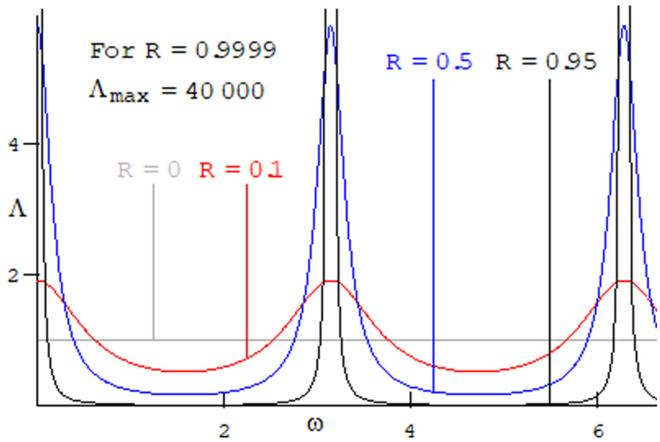

FIG. 2. Examples of the modulation function Λ for various confinement levels, which are determined by the value of R (ω is expressed in units of $\pi c/L$). The $\Lambda_{\max}$ value is calculated from Eq. (5).

formalism of the generalized creation and annihilation operators that follows. The presence of the distribution of Dirac's δ imposes that the operators are no longer dimensionless. Since they describe field-amplitude distributions for an infinitesimal frequency range, these operators are physically meaningful only under proper integration [34,35,40].

To find the relationship between the four operators, allowing us to derive the other commutators for $\hat{a}_{\rm in}^{-}$ and $\hat{a}_{\rm in}^{+}$, we define the relevant boundary conditions. Specifically, we use energy conservation and Stokes's reversibility principle using the amplitude transmittance t and reflectivity r at the microcavity interface. We also note that the field must vanish at the $R = 1$ reflector at $z = -L$. Taking $t = i\sqrt{1-R}$ and $r = -\sqrt{R}$ [36], these conditions are

$$\hat{a}_{\rm in}^{-} = i\sqrt{1-R}\,\hat{a}_{\rm ex}^{-} - \sqrt{R}\,\hat{a}_{\rm in}^{+}, \tag{2a}$$

$$\hat{a}_{\rm ex}^{+} = i\sqrt{1-R}\,\hat{a}_{\rm in}^{+} - \sqrt{R}\,\hat{a}_{\rm ex}^{-}, \tag{2b}$$

$$\hat{a}_{\rm in}^{-}\exp(i\omega L/c) + \hat{a}_{\rm in}^{+}\exp(-i\omega L/c) = \hat{0}. \tag{2c}$$

By writing $\hat{a}_{\rm in}^{-}$ and $\hat{a}_{\rm in}^{+}$ in terms of $\hat{a}_{\rm ex}^{-}$ and using Eq. (1), we find that

$$\left[\hat{a}_{{\rm in},\omega}^{-}, \hat{a}_{{\rm in},\omega'}^{-\dagger}\right] = \left[\hat{a}_{{\rm in},\omega}^{+}, \hat{a}_{{\rm in},\omega'}^{+\dagger}\right] = \Lambda(\omega)\delta(\omega-\omega')\hat{I}, \tag{3}$$

where the modulation function Λ is (see Fig. 2)

$$\Lambda(\omega) = \frac{1-R}{1+R-2\sqrt{R}\cos(2\omega L/c)}. \tag{4}$$

Note the similarity between Eq. (4) and the predicted ratio between the internal and external light intensities for a semiopen cavity of length L [38]. The modulation function thus represents the self-interference of photons in a round trip within the microcavity, which is shorter than the coherence length [36]. Additionally, resonance not only modulates the field amplitudes but also controls the EM noise at resonant frequencies according to Eqs. (3) and (4). For pump frequencies $\omega_{\rm p}$ matching the condition for resonance, the value of the modulation function can be much larger than 1 due to enhanced vacuum fluctuations. This maximum value of Λ is calculated using

$$\Lambda_{\max} = \frac{1+\sqrt{R}}{1-\sqrt{R}} \tag{5}$$

in the event of constructive interference of $\omega_{\rm p}$ virtual photons propagating back and forth in the high-confinement structure.

We are naturally led to the following generalized creation and annihilation operators while fulfilling the conditions of the modulated commutator Eq. (3), as shown in a previous work [23]. They must be used inside the resonator. Incidentally, the $\hat{a}_{\rm in}^{\pm}$ operators are related to the canonical $\hat{a}_{\rm ex}^{\pm}$ operators via Eqs. (2).

Even though the generalized operators apply to every standing-wave Fock state of the universe $|n\rangle$, they describe the photon dynamics inside the resonator. On the other hand, the number of photons inside the microresonator $n_{\rm in}$ is obtained from the ratio of intensities inside and outside the resonator and the dimensions of the system [see Eq. (8)].

The generalized operators are (see the Supplemental Material [41], where $\Lambda = \Lambda(\omega)$)

$$\hat{a}_{\rm in}^{\pm\dagger}|n\rangle = \sqrt{n+\Lambda}|n+\Lambda\rangle \tag{6a}$$

and

$$\hat{a}_{\rm in}^{\pm}|n\rangle = \sqrt{n}|n-\Lambda\rangle. \tag{6b}$$

The first state of eigenmode ω is written as $\sqrt{\Lambda}|\Lambda\rangle = \hat{a}_{\rm in}^{\pm\dagger}|0\rangle$. Since $\hat{a}_{\rm in}^{\pm\dagger}\hat{a}_{\rm in}^{\pm}|n\rangle = n|n\rangle$, Λ does not describe the photon number in a mode but is a measure of its fluctuation, which is increased through confinement. This is a threshold to be overcome in order to detect the smallest amplitude variation, from a classical point of view. Accordingly, from Eq. (6b), one sees that quantum states such as $0 < n < \Lambda$, which correspond to bewildering "negative kets," are linked to eigenmodes that are too buried in vacuum field fluctuations to be clearly identifiable in a classical way and formally describable in terms of harmonic modal functions.

It is clear that the above operators define generalized states, which are not usual number (Fock) states (unless Λ is fortuitously an integer). Rather, they appear as states that should be described by fractional Hermite's polynomials (see Eq. (3.49) in [42] and Eqs. (3.13) and (3.14) in [43]). It is noteworthy that in a quasicontinuous basis space the integral of $\Lambda(\omega)\hat{a}_{\rm in}^{\pm\dagger}|0\rangle$ over frequency introduces a superposition of vacuum states containing information about the total quantum noise level in a specified frequency domain. This might be useful when considering quasimodes.

We stress that n is the number of photons belonging to a universe's eigenmode, not only to a quasimode of the resonator. As expected, the generalized operators recover their canonical form whenever $\Lambda = 1$, as in the case of free space.

Inside a resonator the quantum noise level due to vacuum field fluctuations is spectrally and spatially modulated. The spatial modulation of Λ matches the spatial mode profile of the resonators. In the case of sufficiently high finesse Gires-Tournois or Fabry-Pérot resonators, the modulation can be described by sine functions (since the operators describe the electric fields). In a highly confining resonator ($R \approx 1$) where steady-state conditions are met, the noise level might be so important that inside the resonator at least Λ real photons must be added to or removed from an eigenmode in order to detect a noticeable variation in its electric field amplitude. This is especially true at frequencies close to the resonant modes. This number is only one photon in free space. In other words, for the fields inside the resonator buried in quantum noise, the smallest detectable classical amplitude variation in the corresponding eigenmode is $\sqrt{\Lambda}$, and Λ is the smallest detectable intensity variation. From measurements provided by a "detector" located inside the resonator (the nonlinear medium in our case), the virtual states between $|n\rangle$ and $|n+\Lambda\rangle$ are not physically discernible from one another. This is the case for virtual states between $|n\rangle$ and $|n+1\rangle$ in free space. However, these arguments apply to only a steady-state condition. On a timescale determined by the quality factor of the resonator, the addition (removal) of one input photon at ω outside the resonator, corresponding to Λ photons accumulating in (being expelled from) the resonator, is linked to a final steady state that is reached only asymptotically in time. Of course, a convenient approximation of steady-state conditions is obtained for time much larger than the time constant of the resonator Q/ω. This process requires a total addition (removal) of $\Lambda+1$ photons to the eigenmode.

This discussion is further supported by the calculation of the single-mode degree of second-order coherence $g^{(2)}$. For the forward-propagating component inside the resonator and with τ being the detection time delay, we write [40]

$$g_{\text{in}}^{(2)}(\tau)=\frac{\langle\hat{a}_{\text{in}}^{+\dagger}\hat{a}_{\text{in}}^{+\dagger}\hat{a}_{\text{in}}^{+}\hat{a}_{\text{in}}^{+}\rangle}{\langle\hat{a}_{\text{in}}^{+\dagger}\hat{a}_{\text{in}}^{+}\rangle^2}=g_{\text{in}}^{(2)}(0). \tag{7}$$

With n_{ex} being the number of photons in the rest of the universe ($n_{\text{ex}}+n_{\text{in}}=n$), while assuming that the resonator and the rest of the universe are both nonabsorbing media and α represents the ratio of the refractive index of the resonator over the refractive index of the rest of the universe, we can write (see Eq. (5) in [23])

$$n_{\text{in}}=\alpha\frac{L}{D}\Lambda n_{\text{ex}}=\frac{\alpha L\Lambda}{D+\alpha L\Lambda}n. \tag{8}$$

In terms of linear photon densities, which are more useful for experimental investigations because $D\gg L$, we can write

$$\frac{n_{\text{in}}}{L}=\frac{\alpha\Lambda}{D+\alpha L\Lambda}n\approx\alpha\Lambda\frac{n}{D}. \tag{9}$$

At this point, making use of Eq. (7) and with $(\Delta n)^2$ being the variance, the single-mode degree of second-order coherence inside the resonator can also be generalized to

$$g_{\text{in}}^{(2)}(0)=\frac{\langle\hat{a}_{\text{in}}^{+\dagger}(\hat{a}_{\text{in}}^{+}\hat{a}_{\text{in}}^{+\dagger}-\Lambda)\hat{a}_{\text{in}}^{+}\rangle}{\langle\hat{a}_{\text{in}}^{+\dagger}\hat{a}_{\text{in}}^{+}\rangle^2}=1-\frac{\Lambda}{\langle n\rangle}+\frac{(\Delta n)^2}{\langle n\rangle^2}. \tag{10}$$

Because $(\Delta n)^2$ is a positive value,

$$g_{\text{in}}^{(2)}(0)\geqslant 1-\frac{\Lambda}{\langle n\rangle}. \tag{11}$$

For photon-number Fock states,

$$g_{\text{in}}^{(2)}(0)=1-\frac{\Lambda}{n}. \tag{12}$$

In order to ensure a positive value for $g_{\text{in}}^{(2)}(0)$, the mean number of real photons that are partially buried in the quantum noise constituted of virtual photons must exceed the level Λ, instead of the familiar 1 in free space. In other words, a nonvanishing optical signal occurs only for a superposition of eigenmodes that contain photon-number states with values of n differing by Λ.

In the case of sufficiently low variance $[(\Delta n)^2\ll\langle n\rangle^2]$, a nearly coherent state in the sense $g_{\text{in}}^{(2)}(0)=1$ [not in the Poissonian statistics sense $(\Delta n)^2=\langle n\rangle$] can emerge inside a resonator only when $\langle n\rangle\gg\Lambda$. More generally, making use of the "relative variance" ν (also known as the "Fano factor" [44], sometimes referred inadvertently to as the "Fano parameter" [33], with the latter representing the line-shape asymmetry in the Fano resonance), defined as $\nu\langle n\rangle=(\Delta n)^2$, Eq. (10) is written as

$$g_{\text{in}}^{(2)}(0)=1+\frac{\nu-\Lambda}{\langle n\rangle}. \tag{13}$$

In the same way that, here, ν is a measure of the single-mode field fluctuations due to real photons, inside the resonator Λ is a measure of the single-mode field fluctuations due to virtual photons, which must be overcome to obtain a positive value for $g_{\text{in}}^{(2)}$. Therefore, it appears that in the presence of resonators, the coherence condition $g^{(2)}=1$ is uncoupled from the Poissonian statistics condition $\nu=1$, which is the case in free space. Inside a resonator, the modulation function Λ is interpreted as the threshold variance that a signal must overcome to be distinctive from quantum noise. Remarkably, with the exception of $\langle n\rangle=0$, the photon statistics inside the resonator appear to be coherent (meaning $g^{(2)}=1$) when $\nu=\Lambda$ and even for $\nu>1$. Likewise, the photon statistics inside the resonator appear to be incoherent when $\nu\neq\Lambda$.

One notices that the expected positive value for second-order coherence is disobeyed when $\langle n\rangle<\Lambda$, which produces "negative coherence" (or "superincoherence"). The formal constraints $0\leqslant|g^{(1)}|\leqslant 1$ [45] and $0\leqslant g^{(2)}<\infty$ [40] are deduced only for a coherence time that can be unambiguously defined through an averaging process, which requires long integration times. For experiments that involve a very low number of photons, the coherence time is undetermined. Accordingly, the above constraints are relaxed and in some circumstances could be violated. From Eqs. (10) and (13), one notices that for a sufficiently low variance ν or for a sufficiently high Λ value, $g_{\text{in}}^{(2)}$ is negative. Because the second term in Eq. (13) could be related to an integral form of the Glauber-Sudarshan $P(\alpha)$ function, which is known to eventually have negative values [45] (which can be used as the definition of nonclassical light), the relationship between Eq. (13) and $P(\alpha)$ should be established. Such study is postponed for future work.

Despite recent progress in SHG using incoherent light sources [46–52] and other nonlinear processes [15,53,54], efficient SHG requires coherent light sources. Indeed, the higher the probability is for two real photons to be found in a nonlinear medium at the same position at the same time, the higher the probability is to create a second harmonic photon. Therefore, a high $g_{\text{in}}^{(2)}(0)$, which favors photon bunching, is also favorable to SHG. However, from Eq. (10), Λ has an adverse effect on $g_{\text{in}}^{(2)}$. Even considering an eigenmode with $\langle n\rangle>\Lambda$ such that $g_{\text{in}}^{(2)}$ is positive, $g_{\text{in}}^{(2)}$ decreases as Λ

increases. This can be achieved through a variation of the pump angular frequency ω toward a resonant mode by changing the semireflecting mirror reflectance R or by changing the cavity length L (see Fig. 2). The decreasing rate of $g^{(2)}_{\rm in}(0)$ is especially important for low $\langle n\rangle$ values. Although antibunching is usually defined by $g^{(2)}_{\rm in}(\tau) > g^{(2)}_{\rm in}(0)$ [55–57], here, it is permitted to interpret the decrease in $g^{(2)}_{\rm in}$ as a "structural antibunching," which reduces the SHG efficiency.

Finally, the modulation function Λ is a measure of the minimal level of light intensity fluctuations in an eigenmode inside the resonator. Indeed, it is easily determined that for the forward-propagating component inside the resonator we can write [40]

$$\langle n|\hat{E}^{+2}_{\rm in}|n\rangle = \frac{\hbar\omega}{2\epsilon_o}\left(n+\frac{\Lambda}{2}\right). \tag{14}$$

For a single eigenmode, the ratio of the intensity inside the resonator to the outside determines the relative probability per unit of length P_Λ to detect a photon inside the resonator relative to the rest of the universe. Assuming $\alpha = 1$ for simplicity, we then write

$$P^+_\Lambda = \frac{\langle n|\hat{E}^{+\dagger}_{\rm in}\hat{E}^+_{\rm in}|n\rangle}{\langle n|\hat{E}^{+\dagger}_{\rm ex}\hat{E}^+_{\rm ex}|n\rangle} = \frac{n+\Lambda/2}{n+1/2}. \tag{15}$$

One sees that for an increasing Λ, P_Λ increases proportionally. As expected, this indicates that the photons accumulate into the quasimodes of the resonator. This is associated with the "concomitance law" discussed in [23,37], which establishes an intimate link between vacuum field fluctuations and the density of states. Indeed, this concomitance emphasizes the complementary points of view based of the seemingly diverging concepts of density of states and vacuum field fluctuations. In addition, Eq. (15) gives further physical interpretation to Λ. When $n \gg \Lambda$, the probability is always close to unity, and the effects of confinement vanish. Due to the relatively low value of $\Lambda_{\rm max}$, this could explain why the predictions described in this paper, especially about the eventual existence of a threshold for SHG, have not yet been reported. Experimentally, the use of a low pump level and photon-counting methods appear to be necessary. On the other hand, when n is sufficiently small, Λ is no longer negligible. When the resonator is highly confining (in resonance), then $\Lambda \gg 1$, and $P^+_\Lambda \sim \Lambda$. When the resonator is expelling (out of resonance) when $\Lambda < 1$, this probability is lower than 1. As anticipated, this last result gives a probability of exactly Λ for the zero-point energy state ($n = 0$), which is the case of virtual photons.

Many of the above arguments, especially the threshold variance seen in Eq. (13) that describes the pump-beam coherence inhibition inside the resonator at low pumping level, support the idea that SHG might also be inhibited inside a resonator to the point that a SHG threshold exists. The following sections will theoretically demonstrate that this is formally the case.

III. SECOND-HARMONIC-GENERATION HAMILTONIAN

In order to validate the modulated commutator and the generalized operators, we require a detection system present outside of the microcavity to be able to infer the effects of Eqs. (3) and (4) on an optical processes taking place inside. This is possible by studying how the nonlinear optical process of SHG is modulated by Λ. The microresonator in Fig. 1 is pumped by a laser with an appropriate value of $\omega_{\rm p}$ to match the resonance condition of the microcavity.

Since it is simpler to expend the field operators and perform calculations in terms of a discrete basis of well-defined mode profiles, here, we deviate from our rigorous approach in Secs. I and II and consider the use of quasinormal-mode formalism (i.e., approximation of quasimodes by superposition of very sharp "Lorentz" distribution profiles that ultimately become "Dirac δ" distribution profiles). We stress that this working hypothesis preserves the physical significance of Secs. I and II, especially concerning the Λ modulation function. Accordingly, as a simplification that maintains the usual formal handling of the SHG process without loss of generality from Secs. I and II, in this section we assume a very high quality factor resonator. We can then approximate quasinormal modes as normal eigenmodes. The $2\omega_{\rm p}$ mode, which is populated by the nonlinear effect, is also assumed to be a normal eigenmode.

The Hamiltonian is written as the total energy contained in the EM fields inside the microcavity

$$\hat{H} = \frac{1}{2}\int\left(\epsilon(E_{\rm p})\hat{E}^2 + \frac{\hat{B}^2}{\mu_o}\right)d^3\mathbf{r}, \tag{16}$$

where $\epsilon(E_{\rm p})$ is the permittivity of the nonabsorbing dielectric material ϵ modulated by the harmonic pump field $E_{\rm p}$. Using the approximation for second-order nonlinear Pockels effect in the steady state, an expression for $\epsilon(E_{\rm p})$ is written as

$$\epsilon(E_{\rm p}) = \epsilon_{\rm r}\epsilon_o + \chi^{(2)}_{\rm e}\epsilon_o E_{\rm p}(\mathbf{r})\cos(\omega_{\rm p}t), \tag{17}$$

with $E_{\rm p}(\mathbf{r})$ being the spatial envelope of the pump field and $\chi^{(2)}_{\rm e}$ being the second-order electric susceptibility term.

The creation and annihilation operators for photons are introduced by the electric- and magnetic-field operators through standard (Coulomb gauge) second quantization. The spatial mode profiles $\mathbf{u}_j$ of the jth normal mode (standing-wave basis [58]) corresponding to ω_j are real (sine) functions [45],

$$\hat{E} = \sum_j i\sqrt{\frac{\hbar\omega_j}{2\epsilon}}(\hat{a}_j - \hat{a}^\dagger_j)\mathbf{u}_j, \tag{18a}$$

$$\hat{B} = \sum_j \sqrt{\frac{\hbar}{2\epsilon\omega_j}}(\hat{a}_j + \hat{a}^\dagger_j)\mathrm{rot}\mathbf{u}_j. \tag{18b}$$

Here, the operators $\hat{a}$ and $\hat{a}^\dagger$ refer to those inside the microresonator (the "in" subscripts have been removed for simplicity).

By placing Eq. (17) into Eq. (16), the Hamiltonian can be written in the form $\hat{H} = \hat{H}_o + \hat{H}'$. Taking the nonlinear term in $\epsilon(E_{\rm p})$ as being small compared to $\epsilon_{\rm r}\epsilon_o$, we can use perturbation theory. The first term, $\hat{H}_o$, is the canonical Hamiltonian of a cavity containing EM fields in a linear medium,

$$\hat{H}_o = \sum_j \hbar\omega_j(\hat{a}^\dagger_j\hat{a}_j + \hat{I}/2). \tag{19}$$

The solutions to the temporal evolution of this familiar system are known to be simply harmonic,

$$\hat{a}_j^\dagger(t) = \hat{a}_{j0}^\dagger \exp(i\omega_j t), \tag{20a}$$

$$\hat{a}_j(t) = \hat{a}_{j0} \exp(-i\omega_j t). \tag{20b}$$

The correspondence between the creation and annihilation operators of the jth normal mode and complex exponential functions of angular frequency ω_j is used below to select particular nonlinear processes. The second term, $\hat{H}'$, is the perturbation from the nonlinear optical process inside the microcavity. Calling this term the interaction Hamiltonian, it represents the nonlinear coupling between normal modes,

$$\hat{H}' = -\hbar \cos(\omega_p t) \sum_i \sum_j \kappa_{i,j} (\hat{a}_i - \hat{a}_i^\dagger)(\hat{a}_j - \hat{a}_j^\dagger), \tag{21}$$

where $\kappa_{i,j}$ is the effective coupling constant characteristic of the nonlinear interaction of modes within the microcavity,

$$\kappa_{i,j} = \frac{\chi_e^{(2)} \sqrt{\omega_i \omega_j}}{4\epsilon_r} \int E_p(\mathbf{r}) \mathbf{u}_i \cdot \mathbf{u}_j d^3\mathbf{r}. \tag{22}$$

Phase-matching conditions are assumed for simplicity. Any phase mismatch found in an experimental setup could be described by simply reducing the $\kappa_{i,j}$ value. An appropriate spatial envelope $E_p(\mathbf{r})$ is chosen in order to optimize the $\kappa_{i,j}$ value of interest.

The photonic process of SHG is described as two ω_p photons annihilated to create a single $2\omega_p$ photon. The pump wave ω_p is designated as the $j = 1$ mode, and the SHG signal $2\omega_p$ is designated as the $j = 2$ mode. All other mode-coupling terms in Eq. (21) are ignored. Formally, all $\kappa_{i,j}$ values are approximated to have a null value, either by using an appropriate nonlinear material or by assuming large phase mismatch between waves i and j, with the exception of $\kappa_{1,2} = \kappa_{2,1}$. Only processes satisfying the $\omega_2 = 2\omega_1$ condition are selected to write the SHG interaction Hamiltonian

$$\hat{H}' = \frac{\hbar\kappa}{2} (\hat{a}_1 \hat{a}_1 \hat{a}_2^\dagger + \hat{a}_1^\dagger \hat{a}_1^\dagger \hat{a}_2). \tag{23}$$

For simplicity, $\omega = \omega_p$, and $\kappa = 2\kappa_{1,2}$. Note that we used Euler's formula to write $\cos(\omega t)$ in terms of complex exponential functions, then used Eq. (20) to relate those to creation and annihilation operators. The first term represents the SHG process itself. The second term is the process of degenerate parametric fluorescence, being the adjoint of the first term. Both are present in Eq. (23) to preserve dynamical equilibrium and to ensure that the Hamiltonian remains Hermitian.

IV. EQUATIONS OF MOTION

The temporal evolution of SHG in a high-confinement structure can now be determined while taking into account the effects of the generalized operators through the modulated commutator. Using the SHG Hamiltonian, the solution is obtained by applying Heisenberg's equation of motion

$$\frac{d\hat{a}_j}{dt} = -\frac{i}{\hbar}[\hat{a}_j, \hat{H}]. \tag{24}$$

Under first-order perturbation theory, we take the known result of these equations when $\hat{H} = \hat{H}_o$ as the first term of $d\hat{a}_j/dt$,

$$\frac{d\hat{a}_j}{dt} = -i\omega_j \hat{a}_j - \frac{i}{\hbar}[\hat{a}_j, \hat{H}']. \tag{25}$$

How $\hat{a}_j$ commutes with $\hat{H}'$ includes all effects from the microcavity and acts as a perturbation to the familiar $\hat{H}_o$ system. With Eqs. (23) and (3), we write the equations of motion for the two relevant modes in SHG [37],

$$\frac{d\hat{a}_1}{dt} = -i\omega_1 \hat{a}_1 - i\kappa \Lambda \hat{a}_1^\dagger \hat{a}_2, \tag{26a}$$

$$\frac{d\hat{a}_2}{dt} = -i2\omega_1 \hat{a}_2 - i\frac{\kappa}{2} \Lambda \hat{a}_1 \hat{a}_1. \tag{26b}$$

These equations form the coupled differential equations of motion (CDEMs) that describe the dynamics of the photonic process. When combined with their equivalent equations for $\hat{a}_1^\dagger$ and $\hat{a}_2^\dagger$, the quantum version of the Manley-Rowe relation of SHG can be found:

$$\frac{d(\hat{a}_1^\dagger \hat{a}_1)}{dt} + 2\frac{d(\hat{a}_2^\dagger \hat{a}_2)}{dt} = \hat{0}. \tag{27}$$

This confirms that one ω_2 photon is created simultaneously as two ω_1 photons are annihilated.

A common approximation is to ignore pump depletion. Experimentally, the laser source maintains a near-constant beam containing far more ω_1 photons than the number of ω_2 photons generated by the SHG process. In such a case, the expected number of photons present in the ω_1 resonator quasimode is constant in time and equal to its initial value,

$$n_1(t) = n_{10}, \tag{28}$$

where n_j is the expectation value of $\hat{a}_j^\dagger \hat{a}_j$ and the solutions for $\hat{a}_1^\dagger$ and $\hat{a}_1$ are given by Eq. (20).

Under this approximation, the CDEMs can now easily be solved. Focusing on the mode of interest, the $\hat{a}_2$ equation of motion in Eq. (26) becomes

$$\frac{d\hat{a}_2}{dt} = -i2\omega_1 \hat{a}_2 - i\frac{\kappa}{2} \Lambda \hat{a}_{10} \hat{a}_{10} \exp(-i2\omega_1 t). \tag{29}$$

The solution of this equation allows us to write the expected number of SHG photons in the harmonic cavity quasimode as

$$n_2(t) = n_{20} + n_{10}(n_{10} - \Lambda)\frac{\kappa^2}{4}\Lambda^2 t^2. \tag{30}$$

Often, $n_{20} = 0$ (no injection) is experimentally more relevant. As expected from the arguments exposed in the previous section, the modulation function Λ can be interpreted as a threshold variance that the pump wave must overcome to generate a perceptible SHG signal inside the microresonator. As long as the threshold value is overcome, from the very first moment the photonic system is triggered at $t = 0$, the number of ω_2 photons monotonically increases. However, the approximation of ignoring pump depletion is not valid at times t where $n_2(t)$ is no longer several orders of magnitude smaller than n_{10}. This condition can more easily be met in a steady state with continuous wave pumping but may present additional experimental complications under a pulsed pump setup, especially for ultrashort pulses where the beam is far from being monochromatic.

Additional impacts of the modulation function on the SHG process can be seen in Eq. (30).

The Λ^2 term represents the classical pump-field quadratic-dependence amplification effect of the high-confinement structure. The second-harmonic signal production rate dn_2/dt is stimulated by an increase in vacuum field fluctuations due to resonance. As a numerical example, for $2L/c = 3$ fs and $R = 0.9$, $\Lambda > 30$ near $\omega_p = \pi c/L$ according to Eq. (4), therefore increasing dn_2/dt by three orders of magnitude. This result is a simple consequence of constructive interference within the microcavity.

In the case of a weak pump beam, such that n_{10} is not many orders of magnitude larger than Λ, the Λ^3 dependence represents a nonclassical effect which is specific to the modulation of quantum noise in the system due to confinement. The main impact of the $n_{10} - \Lambda$ term on $n_2(t)$ is how the nonlinear photonic process is triggered at $t = 0$. Because $n_2(t)$ cannot be negative, the minimum number of pump photons to obtain a single ω_2 photon is now increased to $\Lambda + 1$ in a microcavity. The intensification of vacuum field fluctuations hinders the triggering process of SHG in high-confinement systems by increasing the pump threshold level.

As discussed in Sec. II, a coherent pump beam is required for the SHG process to occur. By increasing quantum noise in the microcavity via Λ, incoherence is introduced in the pump wave. Consequently, a greater number of ω_1 photons is required to gain sufficient coherence, which inhibits the triggering of the nonlinear effect. In other words, n_{10} must be greater than the number of virtual photons arising from vacuum field fluctuations in the microcavity to sustain SHG. Note that the ω_1 virtual photons cannot play the role of the pump, as $n_2(t) = 0$ for all Λ if $n_{10} = 0$ in Eq. (30).

In support of the preceding calculations leading to the existence of a threshold for SHG, it is worthwhile to put forward the following alternative approach based on Fermi's golden rule to evaluate the transition probability P. Making use of Eq. (23) and the following eigenstates, where n_{10} and n_{20} are the initial photon numbers in the pump and second-harmonic modes, respectively, with $|i\rangle = |n_{10}, n_{20}\rangle$ and $|f\rangle = |n_{10} - 2\Lambda, n_{20} + \Lambda\rangle$, a simple calculation gives

$$P \propto |\langle f|\hat{H}'|i\rangle|^2 = n_{10}(n_{10} - \Lambda)(n_{20} + \Lambda). \tag{31}$$

One sees that P is positive only for $n_{10} > \Lambda$. Here, too, the modulation function Λ can be interpreted as a threshold intensity that the pump wave must overcome to generate a perceptible and sustainable SHG signal inside the microresonator. It is noteworthy that similar to the free-space condition (where $\Lambda = 1$ and the threshold is $n_{10} = 2$ photons as expected), initial and final states other than the above $|i\rangle$ and $|f\rangle$ lead to $P = 0$.

V. SUMMARY, CONCLUSION, AND PERSPECTIVES

The modulation function Λ for creation and annihilation operators in a microcavity acts as a signature for vacuum field fluctuations. These in turn produce virtual photons capable of affecting nonlinear photonic processes such as SHG. Through high-confinement effects, we have shown that both the SHG signal and the quantum noise are amplified in a microcavity. The production rate of ω_2 photons is inhibited for $n_{10} < \Lambda$, is dependent on Λ^3 for $n_{10} \approx \Lambda$, and is proportional to Λ^2 for $n_{10} \gg \Lambda$. This peculiar signature should help to experimentally validate our theoretical findings. However, the triggering of the SHG process is inhibited by introducing incoherence in the pump wave. Classical theories cannot predict the threshold as it originates purely from quantum noise. Note that by ignoring confinement effects in the microcavity, thus setting $\Lambda = 1$, we recover the familiar quantum description of SHG. In this case, the canonical $n_{10}(n_{10} - 1)$ term in Eq. (30) would produce the two-pump-photon threshold stated by the Manley-Rowe relation. To the best of our knowledge, since the modulation commutator and the ensuing generalized creation and annihilation operators were not previously considered, the results shown in Eqs. (27) and (31) differ from those obtained in the framework of the canonical approach (where Λ is always equal to 1).

The predicted SHG threshold might be difficult to detect experimentally. However, making use of photon-counting methods, experimental investigations are currently in progress. Another experimental challenge arising from our approximations is that the system must be lossless (nonabsorbing) and in a steady state with continuous wave pumping. These conditions may be troublesome to satisfy.

In summary, we have shown that an inclusive understanding of the detailed quantum mechanism is needed in order to study confinement effects in nonlinear photonic systems. Experimental confirmation of both amplification and inhibition effects in SHG would corroborate our result that quantum noise in microcavities can be probed. The experimental confirmation of these quantum effects could open the door to the development of new photonic devices and deepen our fundamental understanding of vacuum field fluctuations.

One of the most important conclusions emerging from our investigation is that a further possible extension of the generalized operators appears to be possible, beyond the case of resonating electromagnetic structures. The modulation function Λ is defined as the "intensity enhancement factor" through the ratio of the intensity inside a resonator I_{in} to the intensity outside the resonator I_{ex} (which is linked to the "field enhancement factor" Λ_E) [23],

$$\Lambda(\omega) = \frac{I_{in}}{I_{ex}} = \left|\frac{E_{in}^{\pm}(\omega)}{E_{ex}^{\pm}}\right|^2 = \Lambda_E^2(\omega). \tag{32}$$

However, it can also be applied to any electromagnetic (nanophotonic) structures that spatially and spectrally alter the electromagnetic field profile not only inside but also in their near surroundings. One can envision that the electromagnetic structure of interest is inserted in a multidimensional plane-parallel capacitor with zero capacitance and subjected to an alternative electric field.

Because vacuum field fluctuations are omnipresent at all frequencies, we assume that (in the Coulomb gauge) a steady-state condition permanently applies to electrical field vacuum fluctuations. Similar to resonance, the spatial distribution profile of the vacuum field fluctuation's intensity and the corresponding field enhancement factor Λ_E can be used to obtain the intensity enhancement factor $\Lambda(\omega)$. Therefore, using the same assumptions to write Eq. (32), we then predict that

$$\Lambda(\mathbf{r},\omega) = \left| \frac{E^{\pm}_{\text{near}}(\mathbf{r},\omega)}{E^{\pm}_{\text{far}}} \right|^2 = \Lambda^2_E(\mathbf{r},\omega). \tag{33}$$

Fortunately, Λ_E can always be determined from classical linear and nonlinear electromagnetism, possibly by involving numerical methods. From Eq. (33), we perceive that the generalized creation and annihilation operators are position dependent, as discussed in [59,60]. Incidentally, because Λ is directly linked to the density of states [23], this last result reinforces the introduction of a local density of states based on the local density of energy, as previously proposed [61,62].

In conclusion, it is our belief that in addition to emphasizing the fundamental importance of vacuum field fluctuations, the present study provides a platform for innovative technological developments. On the other hand, despite the clear picture involved in the generalized operators presented, they remain theoretical and would benefit from experimental validation. In addition to the previously suggested experimental approach to corroborate our findings, here, we suggest some experiments, all conducted using photon-counting methods.

(1) As shown in Eq. (30), at a specific pump frequency the SHG signal inside the resonator depends on Λ^2 and Λ^3. For a highly confining resonator, Λ strongly depends on the incidence angle due to the spectral shifting of the quasimodes [38]. The intensity of the SHG signal emerging from that resonator should also strongly depend on the incidence angle.

(2) We consider a thin nonlinear layer embedded in the microresonator in the form of a Langmuir-Blodgett monolayer molecular film [63]. The presence of a highly confining resonator modulates the vacuum electric field fluctuations as a sine function of its position inside the resonator. Because the SHG signal scales as the square of the pump field, at a constant pumping level the SHG threshold should also be modulated as a function of the location of the thin layer along the cavity axis.

(3) Because the vacuum field fluctuations are present at all frequencies and are assumed to be in a steady-state condition inside all resonators, using a step-function shape pumping, no SHG signal should emerge from the microresonator before a certain time delay, that is, before the number of real photons inside the microcavity has time to overcome Λ (due to the resonator time constant). This delay to detect the SHG signal should depend on R.

(4) Using picosecond-duration light pulses and envisioning a long enough microresonator so that the pulse inside the resonator never overlaps with itself or with other pulses already inside a suitably sized resonator, the SHG signal should depend on the pulse intensity.

(5) In order to avoid the use of delicate photon-counting methods, it would be interesting to resort to recent advances in very high efficiency SHG microring resonators [64,65], which intrinsically reach among the greatest quality factors. The only theoretical requirement is to evaluate Λ inside a toroid cavity, instead of the current Gires-Tournois cavity. Fortunately, Λ_E can always be determined from classical electromagnetism, eventually through numerical methods. On the experimental side, it should suffice to reduce the pump power sufficiently and work as suggested in point 3.

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