Path-integral description of quantum nonlinear optics in arbitrary media

Mosbah Difallah,[1] Alexander Szameit,[2] and Marco Ornigotti[2,3,*]

[1]*Department of Physics, Faculty of Exact Sciences, El Oued University, 39000 El Oued, Algeria*

[2]*Institut für Physik, Universität Rostock, Albert-Einstein-Straße 23, 18059 Rostock, Germany*

[3]*Laboratory of Photonics, Physics Unit, Tampere University, FI-33720 Tampere, Finland*

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We present a method, based on Feynman path integrals, to describe the propagation and properties of the quantized electromagnetic field in an arbitrary, nonlinear medium. We provide a general theory, valid for any order of optical nonlinearity, and we then specialize the case of second-order nonlinear processes. In particular, we show that second-order nonlinear processes in arbitrary media, under the undepleted pump approximation, can be described by an effective free electromagnetic field, propagating in a vacuum, dressed by the medium itself. Moreover, we show that the probability of such processes to occur is related to the biphoton propagator, which contains information about the structure of the medium, its nonlinear properties, and the structure of the pump beam.

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I. INTRODUCTION

Since the early days of laser physics, nonlinear optics has been a very successful and intriguing field of research. Nowadays, with the advent of quantum technologies, nonlinear optics has become the vital part of any quantum optics experiment, as spontaneous parametric down-conversion (SPDC) [1] constitutes the primary source of entangled photons [2]. In recent years, a significant effort has been made to incorporate these sources of entangled photons in integrated on-chip platforms, with the ultimate goal of realizing fully integrated quantum devices which will, ultimately, constitute the basis for quantum computers [3]. To this aim, quantum dots [4], quantum wells [5], photonic waveguides [6–8], plasmonic structures [9], and metamaterials [10–12] have been thoroughly investigated as possible candidates for the next generation of integrated entangled photon sources. The majority of theoretical frameworks currently available to model and design the properties of such systems, however, has been developed mainly for lossless systems (such as optical waveguides), systems made of dispersionless elements only [13–15], or complex geometries, but only interacting with few optical modes [16–18]. An exception to this is represented by a recent work where a method based on Green functions and Born approximation has been proposed to study the nonlinear wave mixing of light fields in metal-dielectric nanostructures of arbitrary geometry [19].

On a seemingly unrelated matter, Feynman path integrals have proven to be a very elegant and successful instrument to describe very complicated systems, ranging from quantum field theoretical problems to many-body problems in condensed matter, and financial markets [20,21]. Although path integrals are especially useful in quantum field theories, they have been introduced into optics a number of times as a way to describe the properties of the electromagnetic field in terms of coherent state representation [22], investigate parametric amplification [23], atom-field interactions beyond the rotating wave approximation [24], quantum decoherence and dephasing in nonlinear spectroscopy [25], and the study of retardation effects and radiative damping [26], to name a few. Moreover, path-integral methods have been used in optics to describe beam propagation [27], optical fiber communications [28], nonparaxial optics [29], and the propagation of the electromagnetic field in homogeneous [30] and inhomogeneous [31] media. In recent years, Bechler [32] has proposed a path-integral approach to describe quantum electrodynamics in linear dispersive media and, recently, a path-integral approach has been proposed to describe temporal dynamics of the quantized electromagnetic field in inhomogeneous media, based on the method of the closed time path generating functional [33].

Here we apply path integrals to study the dynamics of the quantized electromagnetic field in nonlinear, arbitrary media. Such a theory would represent a viable tool to have at one's disposal for analytical (or semianalytical) methods to investigate and quantitatively predict nonlinear interactions of the electromagnetic field in complex systems, such as metamaterials (which are typically realized in terms of stacks of periodically arranged metallic or dielectric structures, with complex geometries, where material loss can no longer, in general, be neglected), epsilon near-zero media (where the dynamics of conduction electrons has to be carefully taken into account), and dielectric media with nontrivial geometries. This is the aim of our work, namely, to develop a path-integral-based theory describing the interactions of the quantized (nonrelativistic) electromagnetic field in an arbitrary, nonlinear medium. We develop a complete theory for the quantized electromagnetic field solely, while the medium, in our analysis, has not been quantized. We leave the full interaction with the quantized electromagnetic field with quantized matter (i.e., photon-polariton interactions) for future works. Here, we focus our attention only on optical nonlinearities,

*marco.ornigotti@tuni.fi

with particular emphasis on second-order processes and, in particular, to SPDC. Our findings show how the cross section for second-order processes can be expressed, within the undepleted pump approximation, in terms of a generalized biphoton propagator, which describes the free propagation of the signal and idler modes in the dressed vacuum of our theory, in the presence of nonlinear interactions.

The purpose of this work is twofold. On one hand, we aim at extending the existing formalism, developed by Bechler [32] for linear systems, to the case of nonlinear media, thus creating a complete framework for the description of the dynamics of the quantized electromagnetic field in arbitrary media. On the other hand, we aim to provide a "universal" framework for the investigation of the properties of the electromagnetic field in nonlinear media. The general approach of path integrals, in fact, will allow us to obtain results that are universally valid for any medium, regardless of its geometry or properties. This, we believe, will constitute a powerful tool to gain insight into complex structures, which are becoming more and more important, in current photonic research and technology development.

This work is organized as follows: in Sec. II, we introduce the formalism of path integrals, which constitutes the main tool used throughout the paper, and we calculate the effective action for an electromagnetic field in a linear, arbitrary medium, in terms of the vector potential solely. This section is largely based on the results obtained in Ref. [32] and has the main goal of fixing the notation and reviewing the method used throughout our paper. The quantization of this effective theory is presented in Sec. III, where a Fourier representation of the dressed photon propagator is also given. In Sec. IV, we discuss the nonlinear interaction of the effective electromagnetic field, while its representation in terms of Feynman diagrams, for the specific case of χ^2 nonlinearity, is given in Sec. V. In Sec. VI, we apply our formalism to some explicit cases, namely, SPDC from a one-dimensional (1D) medium, and the generation of squeezed light by repeated cascaded $\chi^{(2)}$processes. Finally, conclusions are drawn in Sec. VII.

II. PATH-INTEGRAL DESCRIPTION OF THE ELECTROMAGNETIC FIELD IN ARBITRARY MEDIA

In this section, we briefly review the basic formalism developed in Ref. [32], to describe, using the method of path integrals, the propagation of an electromagnetic field in an arbitrary *linear* medium. We start our analysis by considering the following effective partition function:

$$\mathcal{Z}_{\text{eff}}[\mathbf{E},\mathbf{B}] = \int \mathcal{D}\{\mathbf{q}\} e^{\frac{i}{\hbar}S[\mathbf{E},\mathbf{B};\{\mathbf{q}\}]} \equiv e^{\frac{i}{\hbar}S_{\text{eff}}[\mathbf{E},\mathbf{B}]}, \tag{1}$$

where $\mathbf{E} \equiv \mathbf{E}(\mathbf{x}, t)$ $[\mathbf{B} \equiv \mathbf{B}(\mathbf{x}, t)]$ is the electric (magnetic) field, while $\{\mathbf{q}\}$ represents a collection of degrees of freedom associated with matter only (such as, as will be specified later, matter polarization and loss channels). This effective partition function is obtained from the total action of the system, $S[\mathbf{E}, \mathbf{B}, \{\mathbf{q}\}] = \int dt\, d^3x\, \mathcal{L}[\mathbf{E}, \mathbf{B}, \{\mathbf{q}\}]$, with $\mathcal{L}$ being the Lagrangian density, by integrating on all possible configurations of the matter degrees of freedom, collectively described by $\{\mathbf{q}\}$. By doing this, the effect of the medium can be treated macroscopically by means of an effective dielectric constant [32]. Rather than considering the problem in terms of electric and magnetic fields, as is done in Ref. [32], however, here we derive the effective action in terms of the electromagnetic potentials $\mathbf{A} \equiv \mathbf{A}(\mathbf{x}, t)$ and $\Phi \equiv (\mathbf{x}, t)$, which are related to the electric and magnetic fields via the well-known relations [34]

$$\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t} - \nabla\Phi, \tag{2a}$$

$$\mathbf{B} = \nabla \times \mathbf{A}. \tag{2b}$$

The main advantage of this approach is that the effective action can be written in a ready-to-be-quantized form where, however, a suitable gauge needs to be specified. Although there is a very well-known method for including the gauge choice in the path-integral quantization of the electromagnetic field, i.e., the so-called Faddeev-Popov gauge quantization [35], here we explicitly choose a gauge, namely, the Weyl gauge, where $\Phi = 0$ [36], With this gauge choice, we model our system using the Huttner and Barnett Lagrangian density functional [37], which, as a function of the vector potential, can be written as follows:

$$\mathcal{L}[\mathbf{A},\mathbf{P},\mathbf{Y}_\omega] = \mathcal{L}_{em}[\mathbf{A}] + \mathcal{L}_{\text{mat}}[\mathbf{P}] + \mathcal{L}_{\text{res}}[\mathbf{Y}_\omega] + \mathcal{L}_{fm}[\mathbf{A},\mathbf{P}] + \mathcal{L}_{mr}[\mathbf{P},\mathbf{Y}_\omega], \tag{3}$$

where the first three terms are, respectively, the Lagrangian density of the free electromagnetic field, the matter polarization field $\mathbf{P} \equiv \mathbf{P}(\mathbf{x}, t)$, and the reservoir field $\mathbf{Y}_\omega \equiv \mathbf{Y}_\omega(\mathbf{x}, t)$. Their explicit expressions are reported in Appendix A. The interaction of light with matter is described by the term $\mathcal{L}_{fm}[\mathbf{A}, \mathbf{P}]$ and is assumed to be in a minimal coupling form (namely, electric dipole approximation), i.e.,

$$\mathcal{L}_{fm}[\mathbf{A},\mathbf{P}] = -g(\mathbf{x})\,\dot{\mathbf{A}}\cdot\mathbf{P}, \tag{4}$$

where $g(\mathbf{x})$ accounts for the medium geometry. In this model, moreover, the electromagnetic losses are modeled as a reservoir of continuously distributed harmonic oscillators, each characterized by a frequency ω, which interacts only with the matter polarization field via the term

$$\mathcal{L}_{mr}[\mathbf{P},\mathbf{Y}_\omega] = -g(\mathbf{x})\int_0^\infty d\omega\, f(\omega,\mathbf{x})\,\mathbf{P}\cdot\mathbf{Y}_\omega, \tag{5}$$

with $f(\omega, \mathbf{x})$ being the spectral coupling function between the reservoir field and the matter field. The effective action and the effective partition function then becomes

$$\mathcal{Z}_{\text{eff}}[\mathbf{A}] = \int \mathcal{D}\mathbf{P}\mathcal{D}\mathbf{Y}_\omega e^{\frac{i}{\hbar}S[\mathbf{A},\mathbf{P},\mathbf{Y}_\omega]} = e^{\frac{i}{\hbar}S_{\text{eff}}[\mathbf{A}]}, \tag{6}$$

where the integration over $\mathbf{P}$ and $\mathbf{Y}_\omega$ is to be understood as a path integration, and $\mathcal{D}\mathbf{P}$ and $\mathcal{D}\mathbf{Y}_\omega$ are some suitable positive measures defined on an appropriate manifold, which makes the path integration correctly defined [20,38,39]. The details about the calculation of the integrals above are sketched in Appendix B. Using these results, we get

$$S_{\text{eff}}[\mathbf{A}] = S_{em}[\mathbf{A}] + \frac{1}{2}\int dt\, dt'\, d^3x\, g(\mathbf{x})\dot{\mathbf{A}}(t,\mathbf{x})\Gamma(t-t',\mathbf{x})\dot{\mathbf{A}}(t',\mathbf{x}), \tag{7}$$

where the expression for the function $\Gamma(t-t',\mathbf{x})$ is given in Appendix B.

III. QUANTIZATION AND THE EFFECTIVE FREE THEORY

To quantize the above theory, we first insert a coupling term in Eq. (7), which takes into account the interaction of the electromagnetic field with a fictitious source current $\mathbf{J}(t,\mathbf{x})$, and then integrate over all the possible field configurations $\mathbf{A}$. If we then apply the same line of reasoning highlighted in Appendix B to calculate the above integral, we can calculate the above integral, arriving at the following result:

$$\mathcal{Z}_0[\mathbf{J}]=\mathcal{N}_0 e^{\frac{i}{2\hbar}\int dt\,dt'\,d^3x\,d^3x'\,\mathbf{J}(t,\mathbf{x})D(t-t',\mathbf{x}-\mathbf{x}')\mathbf{J}(t',\mathbf{x}')},\quad(8)$$

where $\mathcal{N}_0$ is a suitable normalization constant, which is related to $\mathcal{Z}_0[0]$. Notice, moreover, that the dressed photon propagator $D(t-t',\mathbf{x}-\mathbf{x}')$ is a tensor of rank two, as it connects different components of the vector fields $\mathbf{J}(t,\mathbf{x})$, and $\mathbf{J}(t',\mathbf{x}')$. The knowledge of the quantum partition function $\mathcal{Z}_0[\mathbf{J}]$ is sufficient to derive all the dynamical properties of the field. For example, the propagator can be obtained through functional derivation of $\mathcal{Z}_0[\mathbf{J}]$ with respect to the fields $\mathbf{J}$, i.e., [40],

$$D_{\mu\nu}(t-t',\mathbf{x}-\mathbf{x}')=i\hbar\frac{\delta\mathcal{Z}_0[\mathbf{J}]}{\delta J_\mu(t,\mathbf{x})\delta J_\nu(t',\mathbf{x}')}\bigg|_{\mathbf{J}=0}.\quad(9)$$

The above equation links the μ component of the field at time t and position $\mathbf{x}$, with the ν component of the field, at time t' and position $\mathbf{x}'$, through the dressed photon propagator $D_{\mu\nu}(t-t',\mathbf{x}-\mathbf{x}')$.

Fourier representation of the dressed photon propagator

Equation (9) describes the field dynamics in the time domain. In optics, however, it is easier to work in the frequency domain, as the form of the dielectric function is typically given as a function of frequency, rather than time [34]. It is then useful to find a suitable representation for the dressed photon propagator in the frequency domain, rather than in the time domain. To this aim, we notice that the dressed photon propagator $D(t-t',\mathbf{x}-\mathbf{x}')$ is the Green function of the integrodifferential operator $\hat{R}$, defined in Appendix C. Then, we take the Fourier transform (with respect to time t) of Eq. (C1), call $G_{\mu\nu}(\omega,\mathbf{x})$ the Fourier transform of the dressed propagator, consider only positive frequencies, and use the results of Appendix B to link the Fourier transform of $\Gamma(t-t',\mathbf{x})$ to the dielectric function of the medium $\varepsilon(\omega,\mathbf{x})$. This gives us the following result:

$$\left[\left(-\delta_{\mu\alpha}\nabla^2+\frac{\partial^2}{\partial x_\mu\partial x_\alpha}\right)-\frac{\omega^2}{c^2}\varepsilon(\omega,\mathbf{x})\delta_{\mu\alpha}\right]G_{\alpha\nu}(\omega,\mathbf{x}-\mathbf{x}')=\mu_0\delta_{\mu\nu}\delta(\mathbf{x}-\mathbf{x}').\quad(10)$$

This allows us to interpret $G_{\mu\nu}(x)$ as the Green function of the Helmholtz equation for a monochromatic electromagnetic field, propagating in an arbitrary medium, whose properties are described by the dielectric function $\varepsilon(\omega,\mathbf{x})$ [41]. We can then rewrite Eq. (8), using the Fourier representation $G_{\mu\nu}(\omega,\mathbf{x})$ of the photon propagator, given by the equation above, as follows:

$$\mathcal{Z}_0[\mathbf{J}]=e^{\frac{i}{2\hbar}\int d\omega\,d^3x\,d^3x'\,J_\mu(\omega,\mathbf{x})G_{\mu\nu}(\omega,\mathbf{x}-\mathbf{x}')J_\nu(\omega,\mathbf{x}')}.\quad(11)$$

This is the first result of our work. The dynamics of the electromagnetic field in an arbitrarily shaped, linear medium can be interpreted as those of an effectively free field, propagating in a "vacuum" dressed by the properties of the medium, which define the photon propagator $G_{\mu\nu}(\omega,\mathbf{x})$.

IV. INTERACTING THEORY

The partition function derived above only describes linear dynamics of the electromagnetic field. To include nonlinear dynamics, one has to define a suitable interaction Lagrangian which correctly accounts for the desired nonlinearities. To do that, let us first notice that optical nonlinearities are typically quite small in magnitude and can, therefore, be treated within the framework of perturbation theory. Then, we recall that in optics, nonlinearities enter Maxwell's equation through the field polarization $\mathbf{\Pi}$, which is typically expressed in a power series of the electric field, i.e., $\mathbf{\Pi}=\varepsilon_0(\chi^{(1)}\mathbf{E}+\chi^{(2)}\mathbf{E}^2+\chi^{(3)}\mathbf{E}^3+\cdots)$, where $\chi^{(n)}\equiv\chi^{(n)}(\omega,\mathbf{x})$ is the nth-order susceptibility tensor, with $1+\chi^{(1)}=\varepsilon$ being the dielectric function of the medium [1]. It is not difficult to show that this kind of interaction can be generated by a Lagrangian of the form

$$\begin{aligned}\mathcal{L}_{\text{int}}[\mathbf{A}]&=\sum_{n=2}^{\infty}\frac{(i\omega)^{n+1}}{(n+1)!}\chi^{(n)}\cdot\mathbf{A}^{n+1}\\&=-\frac{i\omega^3}{3!}\chi^{(2)}_{\mu\nu\sigma}A_\mu A_\nu A_\sigma+\frac{\omega^4}{4!}\chi^{(3)}_{\mu\nu\sigma\tau}A_\mu A_\nu A_\sigma A_\tau\\&\quad+\text{higher orders},\end{aligned}\quad(12)$$

where summation over repeated indices has been implicitly understood, and $\{\mu,\nu,\sigma,\tau\}\in\{x,y,z\}$. Using the standard results from Quantum field theory (QFT), we can then write the partition function for the interacting, nonlinear theory as follows [40]:

$$\mathcal{Z}[\mathbf{J}]=\mathcal{N}e^{\frac{i}{\hbar}\int d\omega\,d^3x\,\mathcal{L}_{\text{int}}[\frac{1}{i}\frac{\delta}{\delta\mathbf{J}}]}\mathcal{Z}_0[\mathbf{J}],\quad(13)$$

where $\mathcal{N}$ is a suitable normalization constant, and the argument of the interaction Lagrangian appearing in the exponent above has the meaning of replacing every entry of the vector potential $\mathbf{A}$ with a functional derivative with respect to the correspondent current component [40]. Equation (13) can then be expanded perturbatively. To do that, we assume that the magnitude of each nonlinear susceptibility tensor appearing in Eq. (12) is very small (compared to the linear susceptibility), i.e., $|\chi^{(n)}|\ll\chi^{(1)}$, $\forall n\geqslant 2$, and that the higher-order nonlinearities are progressively smaller, i.e., $|\chi^{(n+1)}|\ll|\chi^{(n)}|,\forall n\geqslant 2$. Under these assumptions, which are verified for typical nonlinear optical materials, we can expand the exponential term appearing in Eq. (13) into a power series to obtain the following result:

$$\mathcal{Z}[\mathbf{J}]=\mathcal{Z}_0[\mathbf{J}]+\sum_{k=2}^{\infty}\mathcal{Z}^{(k)}[\mathbf{J}]+\mathcal{Z}_{\text{cross}}[\mathbf{J}],\quad(14)$$

where $\mathcal{Z}_0[\mathbf{J}]$ is the partition function of the free theory, as given by Eq. (11), $\mathcal{Z}^{(k)}[\mathbf{J}]$ represents the correction to the partition function due to the presence of kth-order nonlinearity in the medium, whose explicit form is given by

$$\mathcal{Z}^{(k)}[\mathbf{J}] = \sum_{n=1}^{\infty} \frac{i^{k+1}}{n!(k+1)!}\left(\frac{i}{\hbar}\right)^n \times \left[\int d\omega\, d^3x\, \omega^{k+1}\, \boldsymbol{\chi}^{(k)} \cdot \left(\frac{1}{i}\frac{\delta}{\delta \mathbf{J}}\right)^{k+1}\right]^n \mathcal{Z}_0[\mathbf{J}], \tag{15}$$

and $\mathcal{Z}_{\rm cross}[\mathbf{J}]$ is the cross nonlinearity term, which contains information about the interplay between the different orders of nonlinearities (i.e., it contains terms proportional to $\Pi_k \boldsymbol{\chi}^{(k)}\boldsymbol{\chi}^{(k+1)}\boldsymbol{\chi}^{(k+2)}\cdots$).

In practical situations, however, this term can be neglected, as it is typically of higher order, with respect to the order of the considered nonlinearity [1]. Equation (14) then represents the most general nonlinear interaction of the electromagnetic field, with a medium containing all orders of optical nonlinearities, each one described by its own nonlinear susceptibility $\boldsymbol{\chi}^{(n)}$. Written in this form, moreover, the above expression can be easily translated in the language of Feynman diagrams. In this work, however, we limit our attention to $\chi^{(2)}-$processes, leaving the more rich structure of $\chi^{(3)}-$nonlinearities to future investigations.

A. Interaction Lagrangian for second-order nonlinear processes

Second-order nonlinear processes involve the interaction of three fields [1]. In this case, then, the nonlinear susceptibility is a rank-3 tensor $\chi_{\sigma\mu\nu}(\omega, \mathbf{x})$ and, according to Eq. (12), the interaction Lagrangian density describing such processes can be written as

$$\mathcal{L}_{\rm int}^{(2)}[\mathbf{A}] = \frac{1}{3!}\chi_{\sigma\mu\nu}(\omega, \mathbf{x})A_\sigma^{(p)}A_\mu^{(s)}A_\nu^{(i)}, \tag{16}$$

where the superscripts $\{p, s, i\}$ stands for pump, signal, and idler photon modes, respectively. Notice, moreover, that in the equation above, the nonlinear susceptibility $\chi_{\sigma\mu\nu}(\omega, \mathbf{x})$ has been redefined in such a way to include the term $-i\omega^3$ appearing in Eq. (12), for later convenience. Equation (16) describes all possible second-order processes, such as second harmonic generation (SHG), sum (difference) frequency generation (SFG/DFG), and (spontaneous) parametric down conversion (SPDC). While SHG consists in the conversion of two degenerate signal and idler modes into a pump one (with SPDC being, practically, its inverse process), SFG (DFG) describes the scattering of a signal (idler) photon into an idler (signal) one, mediated by the presence of a pump photon [1,2].

B. Undepleted pump approximation and the quantum optical dressed vacuum

In practical situations, nonlinear optics experiments are typically carried out by using a very intense pump, which stimulates the onset of nonlinear processes. The reason behind this is very simple: as nonlinear processes are very weak, high intensities, i.e., a high number of photons, are needed in order to make the process probable enough to be observed. Under these working conditions, then, the so-called undepleted pump approximation is used, i.e., the pump mode $A_\sigma^{(p)}$ contains a large number of photons and is often described in terms of coherent states, and typically treated as a classical rather than a quantum field. The occasional conversion of energy from the pump to the signal and idler modes (regulated by the energy conservation constraint $\omega_p = \omega_s + \omega_i$), then, does not affect the number of photons contained in the pump mode, which, in first approximation, can be considered to remain constant. For this reason, therefore, the pump mode is often considered as a classical object and it enters in the dynamics only parametrically, *de facto* contributing to the definition of an effective nonlinear coefficient.

In our theory, we can account for this approximation by promoting the pump field $A_\sigma^{(p)}$ to be a classical field and by introducing the effective nonlinear coupling constant $\lambda_{\mu\nu}(\omega\mathbf{x}) = \chi_{\sigma\mu\nu}(\omega, \mathbf{x})A_\sigma^{(p)}$, so that Eq. (16) can be written as

$$\mathcal{L}_{\rm int}^{(2)}[\mathbf{A}] = \frac{1}{3!}\lambda_{\mu\nu}(\omega, \mathbf{x})A_\mu^{(s)}A_\nu^{(i)}. \tag{17}$$

It is worth noticing that the introduction of the nonlinear interaction, and the consequent undepleted pump approximation, redefine the quantum vacuum of this model in such a way that the true vacuum of the effective theory is dressed by the pump beam and can then be written as $|0\rangle \equiv |\{0_\omega\}_D; \omega_p\rangle$, where $\{0_\omega\}_D$ is a shorthand for describing all the frequency modes of the dressed electromagnetic field and ω_p highlights the fact that the vacuum state is dressed by the pump mode. Within this framework, for example, SPDC can then be described as the spontaneous generation of a signal-idler photon pair from the vacuum, i.e., $|0\rangle \rightarrow |1_s, 1_i\rangle = |\{0_\omega, \cdots 1_{\omega_s}, 1_{\omega_i}, \cdots 0_\omega, \cdots\}; \omega_p\rangle$, where $1_{\omega_{s,i}}$ indicates that a signal (idler) photon has been generated in the mode at frequency $\omega_{s,i}$, respectively, according to the energy-conservation constraint $\omega_s + \omega_i = \omega_p$.

We conclude this section by pointing out an interesting fact. Thanks to the undepleted pump approximation, the interaction Lagrangian appearing in Eq. (17) is quadratic in the vector potential and describes an effective self-interaction of the field, ultimately responsible for $\chi^{(2)}$ nonlinear processes. This hints at the possibility of describing the second-order nonlinear interaction of the electromagnetic field in an arbitrary medium, within the undepleted pump approximation, in terms of the dynamics of a non-Abelian gauge field [42].

C. Partition function for second-order nonlinear phenomena in the undepleted pump approximation

We are now in the position to calculate the explicit expression of the partition function $\mathcal{Z}[\mathbf{J}]$, for $\chi^{(2)}$ nonlinearities in the undepleted pump approximation. To do that, we substitute Eq. (17) into the expression of $\mathcal{Z}_k[\mathbf{J}]$, with $k = 2$, and, for the sake of simplicity, we limit ourselves to consider only the first order of the expansion of the exponential term appearing in Eq. (13). Higher orders, in fact, can be easily derived using the same line of reasoning presented here. With a bit of algebra, it is not difficult to show that the partition function can then be written as $\mathcal{Z}[\mathbf{J}] = \mathcal{Z}_0[\mathbf{J}] + \mathcal{Z}_1[\mathbf{J}] + O(\lambda^2)$, with $\mathcal{Z}_0[\mathbf{J}]$ given by Eq. (11), and $\mathcal{Z}_1[\mathbf{J}]$ can be written, after proper normalization with respect to the free partition function has been taken into account [40] as follows:

$$\frac{\mathcal{Z}[\mathbf{J}]}{\mathcal{Z}_0[\mathbf{J}]} = \left(\frac{1}{2\hbar}\right)^2 \int d^4x d^4y J_\mu(x)\mathcal{X}_{\mu\nu}^{(2)}(x-y)J_\nu(y), \tag{18}$$

where

$$\mathcal{X}^{(2)}_{\mu\nu}(x-y) = \int d^4z G_{\mu\alpha}(x-z)\lambda_{\alpha\beta}(z)G_{\beta\nu}(z-y) \tag{19}$$

is the two-mode (or biphoton) propagator, which describes the dynamics of the signal and idler fields under the effect of the nonlinear interaction.

This is the main result of our work. Second-order nonlinear processes in the undepleted pump approximation are described by the quantum partition function given in Eq. (18), which is in the form of the partition function of a free quantum field, characterized by the biphoton propagator $\mathcal{X}^{(2)}_{\mu\nu}(x-y)$. This result constitutes a generalization of the traditional biphoton wave-function approach to nonlinear optical processes [43–45] since it does not only contain information about the various frequency modes involved in the dynamics, as in the traditional approach, but it also contains information about the spatial distribution of the electromagnetic field inside the medium and the properties of the medium itself.

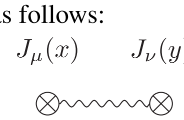
V. FEYNMAN DIAGRAM REPRESENTATION FOR $\mathcal{Z}$[J]

To make our result clearer, we now rewrite Eq. (13) in terms of Feynman diagrams. To start with, we rewrite Eq. (13) in the following form:

$$\mathcal{Z}[\mathbf{J}] = \mathcal{N} e^{\frac{i}{\hbar}\int d\omega d^3x \mathcal{L}_{\rm int}[\frac{1}{i}\frac{\delta}{\delta \mathbf{J}}]}\mathcal{Z}_0[\mathbf{J}]$$
$$= \mathcal{N}\sum_{V=0}^{\infty}\frac{1}{V!}\left\{\frac{i}{3!\hbar}\int d^4z\lambda_{\alpha\beta}(z)\left[\frac{1}{i}\frac{\delta^2}{\delta J_\alpha(z)\delta J_\beta(z)}\right]\right\}^V$$
$$\times \sum_{P=0}^{\infty}\frac{1}{P!}\left[\frac{i}{2\hbar}\int d^4x d^4y J_\mu(x)G_{\mu\nu}(x-y)J_\nu(y)\right]^P. \tag{20}$$

If we compare Eqs. (20) and (18), it is not difficult to see that Eq. (20) reduces to Eq. (18) for $V = 1$ and $P = 2$. This means that Eq. (18) only contains single interaction events ($V = 1$ means, in fact, that only one vertex is allowed in the corresponding Feynman diagram) and two-photon modes, represented by two propagators (hence, $P = 2$) [40].

We can then introduce the Feynman rules for $\chi^{(2)}$ nonlinearities as follows: a dashed line segment represents the dressed vacuum state $|0\rangle \equiv |\{0_\omega\}_D;\omega_p\rangle$; a wiggled line represents the dressed photon propagator $G_{\mu\nu}(x-y)/(i\hbar)$; a dashed crossed dot line indicates an external source current $J_\mu(x)$; a black dot indicates a vertex, where, at maximum, two lines can join (the dashed line representing the dressed vacuum does not count towards this limit). To each vertex representing the nonlinear interaction, the term $(i/3!\hbar)\int d^4z\lambda_{\alpha\beta}(z)$ is associated; at each vertex, energy conservation must be fulfilled. With these rules at hand, we can calculate the partition function $\mathcal{Z}[\mathbf{J}]$ and the related correlation functions $\langle A_\mu(x_1)\cdots A_\nu(x_n)\rangle$ in a very intuitive way. Let us illustrate this with an example. In terms of the Feynman diagrams introduced above, the free partition function $\mathcal{Z}_0[\mathbf{J}]$ can be written as follows:

$Z_0[J] =$  $+$ higher orders, (21)

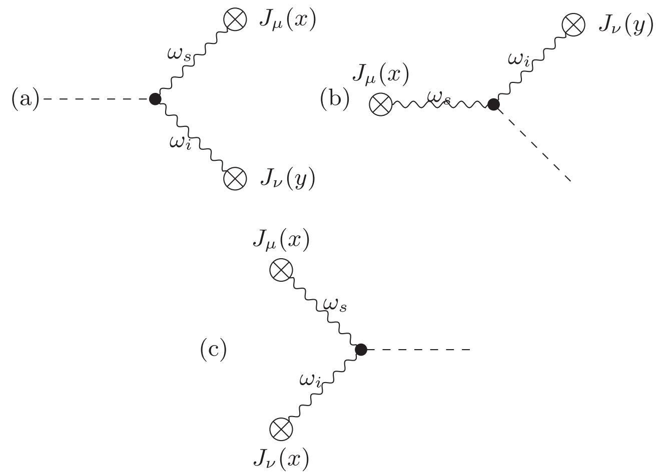
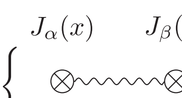
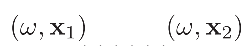


FIG. 1. Relevant Feynman diagrams for $\chi^{(2)}$ processes, corresponding to the various terms composing $\mathcal{Z}[\mathbf{J}]$. (a) Generation of a signal-idler (ω_s)-(ω_i) photon pair from the dressed vacuum (SPDC). (b) Scattering of a photon from the signal mode ω_s to the idler mode ω_i (DFG). (c) Annihilation of a signal-idler photon pair in the dressed vacuum (SFG). The Feynman diagrams depicted in this figure, and all the others throughout the paper, have been generated using TikZ-Feynman [46].

where each current node has been labeled with its corresponding current term, for later convenience. Assume now that we are interested in calculating the two-point correlation function, $\langle A_\mu(x_1)A_\nu(x_2)\rangle$, whose explicit form is given as follows:

$$< A_\mu(\omega,\mathbf{x}_1)A_\nu(\omega,\mathbf{x}_2) >= \left.\frac{\delta^2\mathcal{Z}_0[\mathbf{J}]}{\delta J_\mu(\omega,\mathbf{x}_1)\delta J_\nu(\omega,\mathbf{x}_2)}\right|_{J=0}$$

$= \frac{\delta^2}{\delta J_\mu(\omega,\mathbf{x}_1)\delta J_\nu(\omega,\mathbf{x}_2)}\Big\{$  $\Big\}\Big|_{J=0}$

$=$ 

$= G_{\mu\nu}(\omega,\mathbf{x}_1-\mathbf{x}_2)$. (22)

In terms of Feynman diagrams, this can be understood as follows: every functional derivative in the equation above removes a source (crossed dot) from $\mathcal{Z}_0[\mathbf{J}]$ and labels the correspondent endpoint with the coordinate $x_{1,2} \equiv \{\omega, \mathbf{x}_{1,2}\}$, at which that specific functional derivative is taken. The end result of this calculation is, as expected, the dressed photon propagator $G_{\mu\nu}(\omega,\mathbf{x}_1-\mathbf{x}_2)$.

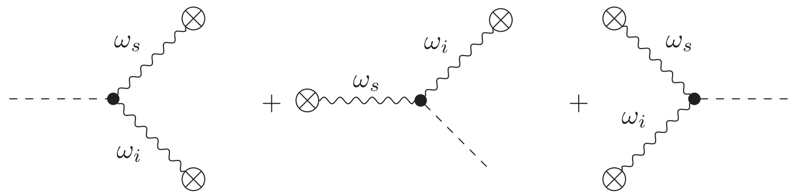
A. Relevant diagrams for $\mathcal{Z}$[J]

The fundamental Feynman diagrams for $\chi^{(2)}$ processes are shown in Fig. 1. There are three relevant diagrams, describing the three basic $\chi^{(2)}$ processes of SPDC [Fig. 1(a)], DFG [Fig. 1(b)], and SFG [Fig. 1(c)]. However, as $\chi^{(2)}$ processes involve three photons, one would expect six different diagrams (as there are $3! = 6$ different ways to arrange the three different diagrams appearing in Fig. 1). The missing three diagrams can be easily obtained from the ones depicted in

Fig. 1 by exchanging the role of the signal and idler modes. Moreover, notice that SHG is a special case of Fig. 1(c), when the signal and idler photons are degenerate, i.e., $\omega_s = \omega_i = \omega_p/2$.

In terms of Feynman diagrams, then, the partition function for $\chi^{(2)}$ processes can be written, at the order $O(\lambda)$, as follows:

$$\mathcal{Z}[\mathbf{J}] =$$



$$+\, \omega_s \leftrightarrow \omega_i \tag{23}$$

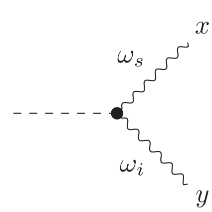
B. Cross section for $\chi^{(2)}$ processes

From a physical point of view, the processes depicted in Fig. 1 are very different. SPDC, for example, is a spontaneous process, originating from the dressed vacuum. DFG and SFG, on the other hand, require the preexisting presence of signal and/or idler photons that can seed the process and make it possible.

Despite the physical and conceptual difference between the various nonlinear processes described by $\chi^{(2)}$ nonlinearities, however, their cross section is the same for any of such processes, and proportional to the two-point correlation function $\langle A_\mu(x) A_\nu(y)\rangle$. The reason for this resides in the fact that in the undepleted pump approximation, the interaction Lagrangian describing $\chi^{(2)}$ processes is quadratic in the vector potential and, therefore, nonlinear interactions can be described in terms of effective, self-interacting free fields, for which the only relevant quantity is the two-point correlation function [47]. For the case of SPDC, for example, we have

$$\sigma_{\text{SPDC}} = < A_\mu(x) A_\nu(y) >= \left.\frac{\delta^2 \mathcal{Z}[\mathbf{J}]}{\delta J_\mu(x) \delta J_\nu(y)}\right|_{\mathbf{J}=0}$$

$$=$$



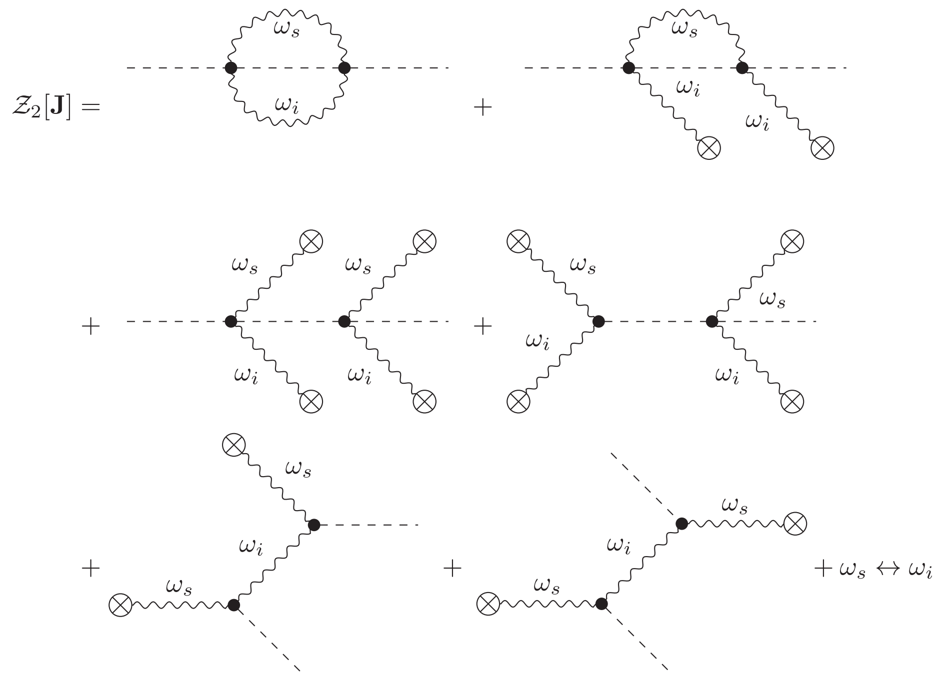
$$= \frac{1}{\hbar^2} \mathcal{X}^{(2)}_{\mu\nu}(x-y)$$

$$= \frac{1}{\hbar^2} \int d^4 z \lambda_{\alpha\beta}(z) G_{\mu\alpha}(x-z) G_{\beta\nu}(z-y), \tag{24}$$

where $x = \{\omega_s, \mathbf{x}\}$, and $y = \{\omega_i, \mathbf{y}\}$.

C. Cascaded $\chi^{(2)}$ processes

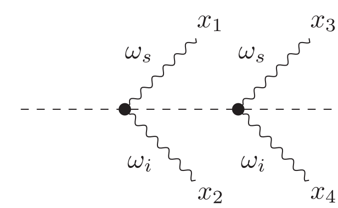
If we want to account for higher-order processes, such as the one depicted in Fig. 2, we need to expand the expression of $\mathcal{Z}[\mathbf{J}]$ so that it also accounts for higher powers of the coupling constant λ. These processes, in optics, are known as cascaded processes. To describe such processes within the framework developed in the previous section, we need to write the partition function as $\mathcal{Z}[\mathbf{J}] = \mathcal{Z}_0[\mathbf{J}] + \mathcal{Z}_1[\mathbf{J}] + \mathcal{Z}_2[\mathbf{J}] + O(\lambda^3)$, where $\mathcal{Z}_2[\mathbf{J}]$ accounts for the occurrence of nonlinear interactions with two vertices. In terms of Feynman diagrams, it can be written as follows:



(25)

Contrary to first-order processes, which all have the same cross section, in this case the cross section is different for different processes. A careful analysis of the diagrams presented above, in fact, reveals that they can be grouped into two groups, namely, those diagrams containing two current sources and those containing four. Processes belonging to these two classes will have different cross sections, as diagrams with only two sources will have an extra factor of two in their cross section (as there are $2! = 2$ equivalent diagrams, corresponding to the two different way to arrange two current sources), while the processes containing four current sources will have an extra factor of 24, as we can arrange the four current sources in $4! = 24$ different ways. As an example, we report the cross section for the cascaded SPDC process, i.e., for the third diagram appearing in Eq. (25),

$$\sigma_{SPDC}^{casc} =$$



$$= \frac{24}{\hbar^4}\mathcal{X}^{(2)}_{\mu\nu}(x_1 - x_2)\mathcal{X}^{(2)}_{\alpha\beta}(x_3 - x_4), \tag{26}$$

where $x_{1,2} = \{\omega_{s,i}, \mathbf{x}_{1,2}\}$ are the coordinates associated to the first signal-idler photon pair, while $x_{3,4} = \{\omega_{s,i}, \mathbf{x}_{3,4}\}$ are associated to the second signal-idler photon pair. In general, $x_{1,2} \neq x_{3,4}$ since the two-photon pair might be generated at a slightly different frequency (especially if a broadband pump is used for this process) or they might be generated in different points inside the nonlinear medium. In the special case, in which the signal and idler modes are degenerate, the above expression simplifies to $\sigma_{\text{SPDC}}^{casc} = 24[\sigma_{\text{SPDC}}]^2$. This result will be useful when describing the generation of squeezed light.

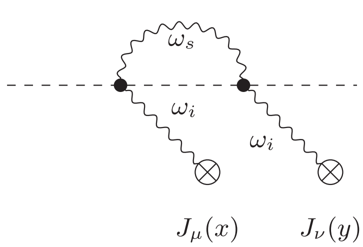


FIG. 2. Higher-order Feynman diagram depicting the process $|0\rangle \to |1_s, 1_i\rangle \to |2_i\rangle$. First, a signal-idler photon pair is created from the dressed vacuum (first interaction point), as described by Fig. 1(a). Then, since the state of the system after this interaction is given by $|1_s, 1_i\rangle$, a second first-order process (second interaction point), namely, the scattering of a signal photon into an idler one [Fig. 1(b)], might take place. However, since this process involves $V = 2$ vertices, it is a process of order $O(\lambda^2)$, and therefore not present in the expression of $\mathcal{Z}[\mathbf{J}]$, which only contains processes up to $O(\lambda)$.

VI. SOME EXAMPLES

In this section, we apply the formalism developed above to two simple examples, namely, the occurrence of SPDC in a one-dimensional, homogeneous, nonlinear medium and the generation of squeezed light from repeated cascaded SPDC processes.

A. SPDC in a one-dimensional waveguide

Let us consider a one-dimensional, homogeneous, nonlinear optical waveguide of length L along the x direction, characterized by a refractive index $n(\omega)$ and a nonlinear susceptibility $\chi^{(2)}_{\mu\nu\sigma} \equiv \chi$. We then introduce the signal and idler wave vectors in the medium, as $k_{s,i}(\omega) = k_0 n_r(\omega_{s,i}) \equiv k_{s,i}$. Then, the nonlinear coupling constant appearing in Eq. (24) assumes a simpler expression, namely, $\lambda_{\alpha\beta}(z) = \chi A_p \exp\left[i(k_p x - \omega_p t)\right]$, and, in particular, is independent of z.

For a one-dimensional, homogeneous medium, the dressed photon propagator can be easily calculated from Eq. (10), giving

$$G(\omega, x-y) = \frac{1}{2ik(\omega)}[\Theta(x-y)e^{i[k(\omega)(x-y)-\omega t]} + \Theta(y-x)e^{-i[k(\omega)(x-y)-\omega t]}], \tag{27}$$

where $\Theta(x)$ is the Heaviside step function [48,49]. We can now calculate the explicit expression of the biphoton propagator $\mathcal{X}^{(2)}_{\mu\nu}(x-y)$, which in this case is given by

$$\mathcal{X}^{(2)}(x-y) = \Theta(x-y)\mathcal{G}(x,y)e^{i\Delta\omega t} L\,\text{sinc}\left(\frac{L\Delta k}{2}\right) + \text{phase mismatched terms}, \tag{28}$$

where $\Delta\omega = \omega_p - \omega_s - \omega_i$ is the frequency mismatch, constrained to be zero by energy conservation, i.e., by $\omega_p = \omega_s + \omega_i$ [1], $\Delta k = k_p + k_s + k_i$ is the phase mismatch, and

$$\mathcal{G}(x,y) = \frac{\chi A_p}{4k_s k_i} e^{-i(k_s x + k_i y)}. \tag{29}$$

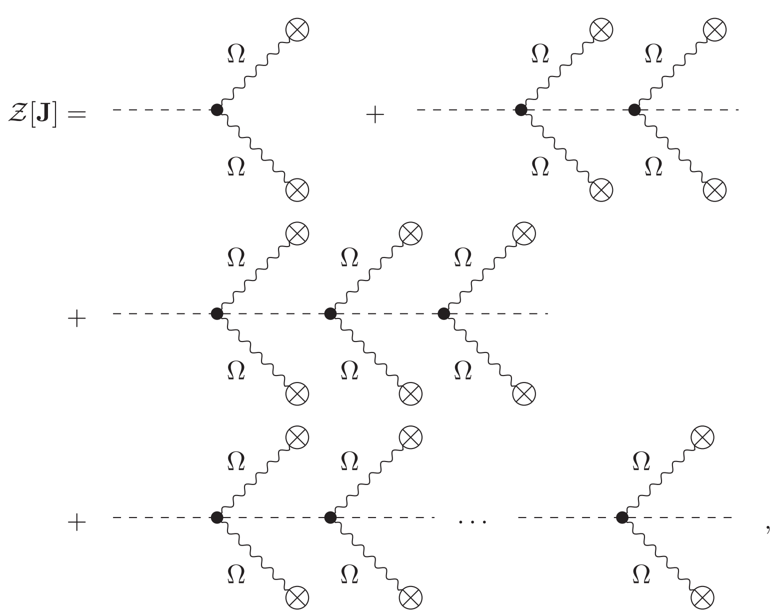
The label *phase mismatched terms* in the above equation, moreover, refers to those terms in the expression of $\mathcal{X}^{(2)}(x-y)$ that violate either the energy or the momentum conservation laws, and that are therefore forbidden. If we assume perfect phase matching, i.e., $\Delta k = 0$, the probability for a SPDC event to occur is then given by

$$P(L) \propto |\sigma_{\text{SPDC}}|^2 \propto L^2\,\text{sinc}^2\left(\frac{L\Delta k}{2}\right), \tag{30}$$

which is in accordance with standard results [1].

B. Squeezing

In this second example, we consider the situation of the occurrence of N cascaded SPDC processes, and we investigate how this process can be connected to squeezing. To make things easier, let us assume that SPDC is degenerate, i.e., that the signal-idler photon pair has the same frequency, namely, $\omega_s = \omega_i \equiv \Omega$, and it is created in the same frequency mode. The partition function describing this process then contains all the SPDC events, up to order N in the expansion in power series of λ, i.e.,



(31)

where the last diagram contains N cascaded SPDC processes. The diagrams shown above describe processes of the type $|0\rangle \to |0\rangle + |2\rangle \to |0\rangle + |2\rangle + |4\rangle \to \cdots \to |0\rangle + |2\rangle + |4\rangle + \cdots + |2N\rangle$.

For cascaded processes, it is not difficult to show that $\sigma^{casc} \propto \sigma^{N/2}$. If we now let $N \to \infty$ and reconstruct the final state of the electromagnetic field as the sum of all these interactions, we obtain

$$|\psi\rangle = \sum_{k\in\{\text{even}\}} \psi_k (\sigma_{\text{SPDC}})^k |k\rangle, \tag{32}$$

where ψ_k is a suitable normalization constant, chosen in such a way that $\langle\psi|\psi\rangle = 1$. A closer inspection on $|\psi\rangle$ reveals that it only contains states with an even number of photons in them. This is the typical form of a single-mode squeezed state [2], i.e.,

$$|\xi\rangle = \sqrt{\text{sech}\, s} \sum_{k\in\{\text{even}\}} \sqrt{\frac{(2k)!}{k!}} \left(-\frac{1}{2}e^{i\theta}\tanh s\right)^k |k\rangle, \tag{33}$$

where $\xi = s\exp(i\theta)$ is the squeezing parameter.

If we compare Eq. (32) with Eq. (33), we can relate, up to a normalization constant, the squeezing parameter ξ with the SPDC cross section and, therefore, with the properties of the nonlinear medium. We then have

$$\psi_k \sigma_{\text{SPDC}}^k = \sqrt{\text{sech}\, s \frac{(2k)!}{k!}} \left(-\frac{1}{2}\right)^k \tanh^k s e^{ik\theta}. \tag{34}$$

If we now call $\sigma_{\text{SPDC}} = \rho_\sigma e^{i\varphi_\sigma}$, where, according to Eq. (24),

$$\rho_\sigma = \frac{1}{\hbar^2} \left|\mathcal{X}_{\mu\nu}^{(2)}(x-y)\right|, \tag{35a}$$

$$\varphi_\sigma = \text{Arg}\left\{\mathcal{X}_{\mu\nu}^{(2)}(x-y)\right\}, \tag{35b}$$

and substitute these expression into Eq. (34), we have, up to a normalization constant,

$$\tanh s = \rho_\sigma = \frac{1}{\hbar^2} \left|\mathcal{X}_{\mu\nu}^{(2)}(x-y)\right|, \tag{36a}$$

$$\theta = \varphi_\sigma = \text{Arg}\left\{\mathcal{X}_{\mu\nu}^{(2)}(x-y)\right\}. \tag{36b}$$

This is an important result. A careful analysis of the equations above reveals, in fact, that the amplitude and phase of the squeezing parameters are, in the general case, not only determined by the properties of the pump beam (as is usually the case [2]), but also by the properties of the medium, encoded in the Green functions for the signal and idler modes, appearing in the definition of the biphoton propagator as well as the geometry of the interaction, i.e., along which directions (with respect, for example, to the principal axes of the nonlinear medium) the signal and idler photons are emitted. For simple cases, the above result reduces to the well-known result that the squeezing parameter is controlled by the pump beam [2]. In fact, if we consider the particular case of one-dimensional SPDC treated above and we assume plane-wave illumination, i.e., $A_p = |A_p| \exp(i\phi_p)$, we can analytically calculate the modulus and phase of the squeezing parameter using Eq. (28), which gives, in the case of perfect phase matching (i.e., $\Delta k = 0$), the following result:

$$s = \ln \sqrt{\frac{4k_s k_i + \chi |A_p| L}{4k_s k_i - \chi |A_p| L}}, \tag{37a}$$

$$\theta = \phi_p - (k_s x + k_i y), \tag{37b}$$

which is in accordance with standard quantum optical calculations [2]. It is interesting to notice, however, that while the squeezing strength s only depends on the pump amplitude, the squeezing phase θ depends on the pump phase and, surprisingly, on the position at which the signal and idler photons are actually detected.

VII. CONCLUSIONS AND OUTLOOK

In conclusion, our work presents a complete toolkit, based on the method of path integrals and Feynman diagrams, for calculating the classical and quantum properties of the electromagnetic field in an arbitrary, nonlinear medium. In particular, we have presented how this method can be used to describe second-order nonlinear processes and that the quantity of interest in this case is the biphoton propagator defined in Eq. (19). Moreover, we have presented two examples of application of our formalism, one to the very simple and well-known case of SPDC from a one-dimensional nonlinear crystal and the other based on the origin of squeezing from multiple cascaded SPDC events.

In future works, we intend to refine this formalism by investigating in more detail the connection between the effective interaction Lagrangian in Eq. (17) and the possibility of describing $\chi^{(2)}$ processes in terms of non-Abelian free gauge fields, with the ultimate goal of defining a suitable framework where the limits and validity of the undepleted pump approximation can be discussed. Moreover, we intend to extend our results to the case of third-order nonlinearities, as well as to include the quantum effects of matter, by studying photon-polariton interactions. The model developed in this work, in fact, already contains information about polaritons in the medium, as has been pointed out already in Ref. [32]. A more detailed study of the interaction, at a quantum level, of photons and polaritons in arbitrary media, moreover, could shine new light on the origin of nonlinear effects in complex media, such as metamaterials and metasurfaces.

ACKNOWLEDGMENTS

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APPENDIX A: LAGRANGIAN DENSITIES OF THE FREE FIELDS

In this Appendix, we report the explicit expressions for the free terms of the Lagrangian density appearing in Eq. (3), namely, the Lagrangian density of the free electromagnetic field,

$$\mathcal{L}_{em}[\mathbf{A}] = \frac{\varepsilon_0}{2}\dot{\mathbf{A}}^2 - \frac{1}{2\mu_0}(\nabla \times \mathbf{A})^2, \tag{A1}$$

the Lagrangian density of the free matter polarization field,

$$\mathcal{L}_{\text{mat}}[\mathbf{P}] = \frac{g(\mathbf{x})}{2\varepsilon_0 \omega_0^2 \beta(\mathbf{x})}\left[\dot{\mathbf{P}}^2 - \omega_0^2 \mathbf{P}^2\right], \tag{A2}$$

and the Lagrangian density of the reservoir field,

$$\mathcal{L}_{\text{res}}[\mathbf{Y}_\omega] = g(\mathbf{x}) \int_0^\infty d\omega\, \frac{\rho(\mathbf{x})}{2}\left[\dot{\mathbf{Y}}_\omega^2 - \omega^2 \mathbf{Y}_\omega^2\right]. \tag{A3}$$

In the above equations, the dot indicates derivation with respect to time. Notice, moreover, that the matter polarization field is modeled by a harmonic oscillator with resonant frequency ω_0. The coefficient $\beta(\mathbf{x})$ is dimensionless and represents the static polarizability of the medium. The quantity $\rho(\mathbf{x})$ appearing in $\mathcal{L}_{\text{res}}$, moreover, is the mass density per unit frequency associated with each reservoir oscillator.

APPENDIX B: DERIVATION OF THE EFFECTIVE PARTITION FUNCTION

In this Appendix, we will show how to calculate the integrals appearing in Eq. (6), using simple arguments, based on Gaussian integrals. Before proceeding with the integration, however, let us rewrite Eq. (6) in the following form, which will be easier to deal with in the next sections:

$$\mathcal{Z}_{\text{eff}}[\mathbf{A}] = e^{\frac{i}{\hbar}S_{em}[\mathbf{A}]}\mathcal{I}_P[\mathbf{A}], \tag{B1}$$

where

$$\mathcal{I}_P[\mathbf{A}] = \int \mathcal{D}\mathbf{P}\, e^{\frac{i}{\hbar}(S_{\text{mat}}[\mathbf{P}]+S_{mf}[\mathbf{A},\mathbf{P}])}\mathcal{I}_Y[\mathbf{P}], \tag{B2}$$

and

$$\mathcal{I}_Y[\mathbf{P}] = \int \mathcal{D}\mathbf{Y}_\omega\, e^{\frac{i}{\hbar}(S_{\text{res}}[\mathbf{Y}_\omega]+S_{mr}[\mathbf{Y}_\omega,\mathbf{P}])}, \tag{B3}$$

and the various actions defined above are defined according to the definitions of the correspondent Lagrangian densities defined in Eq. (3).

1. Calculation of $\mathcal{I}_Y[\mathbf{P}]$

To calculate $\mathcal{I}_Y[\mathbf{P}]$, we need to write the exponent in Eq. (B3), i.e.,

$$\begin{aligned} S_{\text{res}}[\mathbf{Y}_\omega] + S_{mr}[\mathbf{Y}_\omega,\mathbf{P}] &= \int dt\, d^3x \int_0^\infty d\omega\, g(x) \\ &\times\left[\frac{\rho}{2}\dot{\mathbf{Y}}_\omega^2 - \frac{\rho\omega^2}{2}\mathbf{Y}_\omega^2 - f(\omega)\mathbf{P}\cdot\dot{\mathbf{Y}}_\omega\right], \end{aligned} \tag{B4}$$

as a quadratic form, i.e., $(\mathbf{Y}_\omega, \hat{A}\mathbf{Y}_\omega)$. To bring the above term in the desired form, we can first integrate by parts, with respect to time, the last term to shift the time derivative from the reservoir field to the matter field. Then, we can transform the first term by using the identity

$$\left(\frac{\partial\phi}{\partial t}\right)^2 = \frac{\partial}{\partial t}\left(\phi\frac{\partial\phi}{\partial t}\right) - \phi\frac{\partial^2\phi}{\partial t^2}, \tag{B5}$$

and then integrate once more by parts, with respect to time. We can then rearrange the result to obtain

$$\begin{aligned} S_{\text{res}}[\mathbf{Y}_\omega] &+ S_{mr}[\mathbf{Y}_\omega,\mathbf{P}] \\ &= \int dt\, dt'\, d^3x \int_0^\infty d\omega\, g(x) \\ &\times\left\{-\frac{1}{2}[\mathbf{Y}_\omega(t'), \hat{A}(t,t')\mathbf{Y}_\omega(t)] - [\mathbf{b}(t'), \mathbf{Y}_\omega(t)]\right\}, \end{aligned} \tag{B6}$$

where a second time integration has been included to express the operator $\hat{A} = \hat{A}(t', t)$, so that it can be interpreted as an actual propagator (or Green function) and

$$\hat{A} \rightarrow \frac{i\rho g(x)}{2\hbar}\left(\frac{\partial^2}{\partial t^2} + \omega^2\right)\delta(t-t'), \tag{B7a}$$

$$\mathbf{b} \rightarrow -\frac{i}{\hbar}g(x)f(\omega)\delta(t-t')\dot{\mathbf{P}}. \tag{B7b}$$

We are now in the position to solve the integral in Eq. (B3), which gives

$$\mathcal{I}_Y[\mathbf{P}] = \mathcal{N}_Y e^{\frac{1}{2}(\mathbf{b},\hat{A}^{-1}\mathbf{b})}, \tag{B8}$$

where $\mathcal{N}_Y$ is a normalization constant and

$$\begin{aligned} (\mathbf{b}, \hat{A}^{-1}\mathbf{b}) = -\frac{i}{\hbar}\Bigg[&\int dt\, d^3x \int_0^\infty d\omega\, \frac{|f(\omega)|^2 g(x)}{\rho}\mathbf{P}(t)^2 \\ &+ \int dt\, dt'\, d^3x\, \frac{g(x)}{\rho}\mathbf{P}(t)\mathcal{G}(t-t',x)\mathbf{P}(t')\Bigg], \end{aligned} \tag{B9}$$

where we have defined

$$\mathcal{G}(t-t',x) = \int_0^\infty d\omega\, \omega^2|f(\omega)|^2 D_F(t-t',\omega) \tag{B10}$$

as the time-domain Green function of the reservoir field. Notice that the inverse operator $\hat{A}^{-1}$ appearing above can be calculated from the following solution of the one-dimensional wave equation:

$$\left(\frac{\partial^2}{\partial\tau^2} + \omega^2\right)D_F(\tau,\omega) = \delta(\tau), \tag{B11}$$

where

$$D_F(t-t',\omega) = \int \frac{d\Omega}{2\pi}\, \frac{e^{i\Omega(t-t')}}{\omega^2-\Omega^2} \tag{B12}$$

is the Feynman propagator [40]. With this in mind, it is then not difficult to show that

$$\hat{A}^{-1} \rightarrow \frac{\hbar}{i\rho g(x)}D_F(t-t',\omega). \tag{B13}$$

2. Calculation of $\mathcal{I}_P[\mathbf{A}]$

We can now turn our attention to the integral in Eq. (B2), given the results we obtained above for $\mathcal{I}_Y[\mathbf{P}]$. The method to solve this integral is pretty much the same as the one outlined above, i.e., in the form of a Gaussian integral. To this aim, let us first notice that the exponent of Eq. (B8) contains a term proportional to $\mathbf{P}^2$, which can be summed with the correspondent quadratic term appearing in the free part of the matter action, $S_{\text{mat}}[\mathbf{P}]$. To do this, we first define the quantity $v(\omega) = f(\omega)\sqrt{\varepsilon_0\omega_0^2\beta\rho}$, and introduce the scaled resonance frequency,

$$\tilde{\omega}_0^2 = \omega_0^2 + \int_0^\infty d\omega\, \frac{|v(\omega)|^2}{\rho^2}. \tag{B14}$$

The next step is then to integrate by parts the term proportional to $\dot{\mathbf{P}}^2$ and introduce an extra time integration so that the total exponent appearing in the integral (B2) can then be written as $(\mathbf{P}, \hat{A}\mathbf{P}) + (\mathbf{b}, \mathbf{P})$, where, in this case,

$$\hat{A} \rightarrow \frac{ig(x)}{\hbar\varepsilon_0\omega_0^2\beta}\left(\frac{\partial^2}{\partial t^2} + \tilde{\omega}_0^2\right)\delta(t-t') - \frac{ig(x)}{\hbar\rho}\mathcal{G}(t-t',x), \tag{B15}$$

$$\mathbf{b} = -\frac{ig(x)}{\hbar}\dot{\mathbf{A}}. \tag{B16}$$

By now carrying out Gaussian integration over the matter degrees of freedom $\mathbf{P}$, we get the following result:

$$\mathcal{I}_P[\mathbf{A}] = e^{\frac{i}{2\hbar}\int dt\, dt'\, d^3x\, g(x)\dot{\mathbf{A}}(x,t)\Gamma(t-t',x)\dot{\mathbf{A}}(x,t')}, \tag{B17}$$

where $\Gamma(t-t',x)$ is the solution of the following, integrodifferential equation [32]:

$$\frac{1}{\varepsilon_0\omega_0^2\beta}\left(\frac{\partial^2}{\partial t^2}+\tilde{\omega}_0^2\right)\Gamma(t-t',x) - \frac{1}{\rho}\int d\tau\,\mathcal{G}(t-\tau,x)\Gamma(\tau-t,x)=\delta(t-t'). \quad \text{(B18)}$$

3. Physical meaning of $\Gamma(t-t',x)$

The function $\Gamma(t-t',x)$ defined above contains all the information about the medium in which the electromagnetic field is propagating and can be interpreted as a kind of effective dielectric constant for the "dressed" electromagnetic field, as discussed in detail in Ref. [32] and reported briefly in this Appendix.

The effective Lagrangian associated with the effective action defined in Eq. (7) can be written in terms of the electric and magnetic fields in the Fourier domain as follows:

$$\mathcal{L}_{\rm eff}[\mathbf{A}] = \varepsilon_0|\mathbf{E}(\Omega,x)|^2+\frac{1}{\mu_0}\mathbf{B}(\Omega,x)|^2 + g(x)\Omega^2\tilde{\Gamma}(\Omega,x)|\mathbf{E}(\Omega,x)|^2, \quad \text{(B19)}$$

where $\tilde{\Gamma}(\Omega,x)$ is the Fourier transform of $\Gamma(t-t',x)$. According to standard field theory [50], the electric displacement $\mathbf{D}(\Omega,x)$ can be directly derived from the effective Lagrangian as follows:

$$\mathbf{D}(\Omega,x)=\frac{\partial\mathcal{L}_{\rm eff}}{\partial\mathbf{E}^*}=\varepsilon_0\mathbf{E}(\Omega,x)+g(x)\tilde{\Gamma}(\Omega,x)\mathbf{E}(\Omega,x). \quad \text{(B20)}$$

From the above equation and by recalling the constitutive relation $\mathbf{D}=\varepsilon_0\varepsilon\mathbf{E}$ [34], it is possible to define the (positive-frequency) effective dielectric constant as

$$\varepsilon_+(\Omega,x)=1+\frac{g(x)}{\varepsilon_0}\tilde{\Gamma}(\Omega,x), \quad \text{(B21)}$$

while the negative-frequency part of the dielectric constant can be obtained by analytic continuation, namely, $\varepsilon_-(\Omega,x)=\varepsilon_+^*(\Omega,x)$.

APPENDIX C: EXPLICIT EXPRESSION OF THE $\hat{R}$ OPERATOR

The differential operator $\hat{R}$ introduced in Sec. III to express the action $S_q[\mathbf{A},\mathbf{J}]$ in a quadratic form is a vector operator, which can be written as $\hat{R}=\hat{R}^{(0)}-+g(x)\partial^2\hat{\Gamma}/\partial t^2$, where $\hat{\Gamma}$ is the operator, whose Green function is given by $\Gamma(t-t',x)$, as defined in Eq. (B18), and $\hat{R}^{(0)}$ is a vectorial operator, whose components $\hat{R}^{(0)}_{\mu\nu}$ are given as follows:

$$\hat{R}^{(0)}_{\mu\nu}(t-t',x-x')=\left[\left(\varepsilon_0\frac{\partial^2}{\partial t^2}-\frac{1}{\mu_0}\nabla^2\right)\delta_{\mu\nu}+\frac{1}{\mu_0}\frac{\partial}{\partial x_\mu}\frac{\partial}{\partial x_\nu}\right] \delta(t-t')\delta(x-x'), \quad \text{(C1)}$$

where $(\mu,\nu)\in\{1,2,3\}$ and we set $\{x,y,z\}\equiv\{x_1,x_2,x_3\}$.

[1] R. W. Boyd, *Nonlinear Optics*, 3rd ed. (Academic, Cambridge, Massachusetts, 2008).
[2] R. Loudon, *The Quantum Theory of Light*, 3rd ed. (Oxford Science, Oxford, 2000).
[3] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge Unversity Press, Cambridge, 2010).
[4] A. Dousse, J. Suffczynski, A. Beveratos, O. Krebs, A. Lemaitre, I. Sagnes, J. Bloch, P. Voisin, and P. Senellart, Nature (London) **466**, 217 (2010).
[5] A. Nevet, N. Berkovitch, A. Hayat, P. Ginzburg, S. Ginzach, O. Sorias, and M. Orenstein, Nano Lett. **10**, 1848 (2010).
[6] J. W. Silverstone, D. Bonneau, K. Ohira, N. Suzuki, H. Yoshida, N. Iizuka, M. Ezaki, C. M. Natarajan, M. G. Tanner, R. H. Hadfield, V. Zwiller, G. D. Marshall, J. G. Rarity, J. L. O'Brien, and M. G. Thompson, Nat. Photon. **8**, 104 (2014).
[7] H. Jin, F. M. Liu, P. Xu, J. L. Xia, M. L. Zhong, Y. Yuan, J. W. Zhou, Y. X. Gong, W. Wang, and S. N. Zhu, Phys. Rev. Lett. **113**, 103601 (2014).
[8] A. S. Solntsev, F. Setzpfandt, A. S. Clark, C. W. Wu, M. J. Collins, C. Xiong, A. Schreiber, F. Katzschmann, F. Eilenberger, R. Schiek, W. Sohler, A. Mitchell, C. Silberhorn, B. J. Eggleton, T. Pertsch, A. A. Sukhorukov, D. N. Neshev, and Y. S. Kivshar, Phys. Rev. X **4**, 031007 (2014).
[9] M. Kauranen and A. Zayats, Nat. Photon. **6**, 737 (2012).
[10] A. K. Popov and V. M. Shalaev, Appl. Phys. B **84**, 131 (2006).
[11] H. Suchowski, K. O'Brien, Z. J. Wong, A. Salandrino, X. B. Yin, and X. Zhang, Science **342**, 1223 (2013).
[12] D. de Ceglia, M. A. Vincenti, S. Campione, F. Capolino, J. W. Haus, and M. Scalora, Phys. Rev. B **89**, 075123 (2014).
[13] J. A. Crosse and S. Scheel, Phys. Rev. A **83**, 023815 (2011).
[14] M. Kamandar Dezfouli, M. M. Dignam, M. J. Steel, and J. E. Sipe, Phys. Rev. A **90**, 043832 (2014).
[15] D. A. Antonosyan, A. S. Solntsev, and A. A. Sukhorukov, Phys. Rev. A **90**, 043845 (2014).
[16] L. G. Helt, M. J. Steel, and J. E. Sipe, New J. Phys. **17**, 013055 (2015).
[17] L. G. Helt and M. J. Steel, Opt. Lett. **40**, 1460 (2015).
[18] T. Onodera, M. Liscidini, J. E. Sipe, and L. G. Helt, Phys. Rev. A **93**, 043837 (2016).
[19] A. N. Poddubny, I. V. Iorsh, and A. A. Sukhorukov, Phys. Rev. Lett. **117**, 123901 (2016).
[20] H. Kleinert, *Path Integrals in Quantum Mechanics, Statistics, Polymer Physics, and Financial Markets*, 5th ed. (World Scientific, Singapore, 2009).
[21] R. P. Feynman and A. R. Hibbs, *Quantum Mechanics and Path Integrals* (McGraw-Hill, New York, 1965).
[22] M. Hillery and M. S. Zubairy, Phys. Rev. A **26**, 451 (1982).
[23] M. Hillery and M. S. Zubairy, Phys. Rev. A **29**, 1275 (1984).
[24] K. Zaheer and M. S. Zubairy, Phys. Rev. A **37**, 1628 (1988).
[25] Y. Tanimura and S. Mukamel, Phys. Rev. E **47**, 118 (1993).

[26] V. Chernyak and S. Mukamel, J. Chem. Phys. **100**, 2953 (1994).
[27] S. R. Vatsya, J. Opt. Soc. Am. B **22**, 2512 (2005).
[28] A. V. Reznichenko, I. S. Terekhov, and S. K. Turitsyn, J. Phys.: Conf. Ser. **826**, 012026 (2017).
[29] M. C. Braidotti, C. Conti, M. Faizal, S. Dey, L. Alasfar, S. Alsaleh, and A. Ashour, Europhys. Lett. **124**, 44001 (2018).
[30] Y. Dimant and S. Levit, J. Opt. Soc. Am. B **27**, 899 (2010).
[31] J. I. Gersten and A. Nitzan, J. Opt. Soc. Am. B **4**, 293 (1987).
[32] A. Bechler, J. Mod. Opt. **46**, 901 (1999).
[33] A. E. Rubio Lopeza and F. C. Lombardo, Eur. Phys. J. C **75**, 93 (2015).
[34] J. D. Jackson, *Classical Electrodynamics*, 3rd ed. (Wiley, New York, 1998).
[35] L. D. Faddeev, *Gauge Fields: An Introduction To Quantum Theory*, 2nd ed. (Westview, Boulder, Colorado, 1993)
[36] M. Creutz, Ann. Phys. **117**, 471 (1979).
[37] B. Huttner and S. M. Barnett, Phys. Rev. A **46**, 4306 (1992).
[38] J. Iliopoulos, C. Itzykson, and A. Martin, Rev. Mod. Phys. **47**, 165 (1975).
[39] P. D. Drummon and M. Hillery, *The Quantum Theory of Nonlinear Optics* (Cambridge University Press, Cambridge, 2014).
[40] M. Srednicki, *Quantum Field Theory* (Cambridge Unversity Press, Cambridge, 2007).
[41] W. Vogel and D. G. Welsch, *Quantum Optics* 3rd ed. (Wiley, New York, 2006).
[42] P. H. Frampton, *Gauge Field Theories* (Wiley-VCH, Weinheim, Germany, 2008).
[43] F. A. Beduini, J. A. Zielinska, V. G. Lucivero, and Y. A. de Icaza Astiz, Phys Rev. Lett. **113**, 183602 (2014).
[44] M. V. Fedorov, A. A. Sysoeva, S. V. Vintskevich, and D. A. Grigoriev, Las. Phys. Lett. **15**, 035206 (2018).
[45] F. Lenzini, A. N. Poddubny, J. Titchener, P. Fisher, A. Boes, S. Kasture, B. Haylock, M. Villa, A. Mitchell, A. S. Solntsev, A. A. Sukhorukov, and M. Lobino, Light Sci. Appl. **7**, 17143 (2018).
[46] J. Ellis, Comput. Phys. Commun. **210**, 103 (2017).
[47] A. Das, *Field Theory A Path Integral Approach* (World Scientific, Singapore, 2006).
[48] F. W. J. Olver, D. W. Lozier, R. F. Boisvert, and C. W. Clark, *NIST Handbook of Mathematical Functions* (Cambridge University Press, Cambridge, 2010).
[49] F. W. Byron and R. W. Fuller, *Mathematics of Classical and Quantum Physics* (Dover, New York, 1992).
[50] L. D. Landau, and E. M. Lifchitz, *Classical Theory of Fields* (Butterworth Heinemann, Oxford, 1996).