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Quantum theory for light propagation in a nonlinear effective medium

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Starting with the canonical quantization procedure for the electromagnetic field inside an effective (linear or nonlinear) medium, we present a direct-space formulation of the theory of quantum optics. This approach does not use the conventional modal decomposition of the field, but relies on the electromagnetic momentum operator defined in terms of local electric- and magnetic-field operators. The momentum operator contains all the information on the spatial characteristics of the field, and can describe the translation of a short light pulse in a nonlinear medium, without a modal analysis of the pulse. Propagation is described through an operatorial wave equation that relates the temporal evolution of an electromagnetic pulse to its spatial progression. Through this equation, the direct-space approach to quantum optics can treat traveling-wave nonlinear-optical phenomena and, at the same time, account for their quantum statistics. The theory is applied to squeezed-light generation by the parametric down-conversion of a short laser pulse, as an illustration.

I. INTRODUCTION

The classical theory of optics can address the problem of propagation of a short light pulse through a transparent linear or nonlinear medium in a relatively simple way, owing mainly to two features in its formalism. First, the material is considered as a continuous dielectric characterized by a set of phenomenological constants, the optical susceptibilities, and second, the spatial progression of light pulses is calculated explicitly by reducing the electromagnetic wave equation directly into a spatial differential equation for the electric field of the propagating pulse. By contrast, the rigorous theory of quantum optics [1] gives rise to a relatively cumbersome (albeit accurate) formulation of propagative optical phenomena, because of two corresponding features in its basic assumptions. First, the material system is introduced in the form of point charges (or atoms) interacting with the field and, second, all calculations are carried out in reciprocal space in terms of modal (photon) operators that represent electromagnetic excitations delocalized throughout the cavity of quantization.

An important simplification of quantum optics results when the microscopic description of the material, in terms of individual atoms, is replaced by a macroscopic description, in terms of an effective (linear or nonlinear) polarization, analogous to that of classical optics. This constitutes, essentially, an approximation that neglects the field statistics which may result from correlations among the excited atoms in the medium and is therefore valid only at frequencies far from the atomic resonances or atomic ionization frequencies, in the transparency region of the material, where the scattering of the field by the atoms is elastic. In spite of the phenomenological treatment of the medium, such an effective theory still permits a quantum-mechanical description of the field, in the sense that it can treat all the problems associated with the noncommutativity of the field operators (such as spontaneously initiated nonlinear processes or photon statistics) without requiring the introduction of external fluctuations as in the theories in which the field is treated classically.

Several authors have examined the effective theory of quantum optics. The quantization of the electromagnetic field in a homogeneous linear (refractive) dielectric was studied quite early by Jauch and Watson [2]. Later, Shen introduced a procedure that became quite popular in quantum optics, whereby the linear polarization is incorporated in the definition of the field modes [3]. More recently, Glauber and Lewenstein [4] examined the canonical field quantization in an inhomogeneous linear medium (i.e., with a position-dependent dielectric function) in view of an analysis of the quantum-mechanical fluctuation properties of the field inside dielectrics. The quantization of the electromagnetic field in a homogeneous nonlinear medium has been discussed by Hillery and Mlodinow [5] and by Drummond and Carter [6]. Starting with the canonical procedure of quantization, these authors showed that, inside an effective nonlinear medium, the definition of photon creation and annihilation operators (as well as the modal expansion of the Hamiltonian) must be done in terms of the modes of the displacement field D and the vector potential A, since these are the canonical conjugate variables. Modal expansions of the electric field E (such as those often introduced heuristically in nonlinear quantum optics) lead to equations of motion that are not compatible with the macroscopic Maxwell equations.

The second feature of the conventional theory of quantum optics, that is, the use of modes and the reciprocal-space representation, has been adopted essentially because it simplifies the treatment of the quantized free field or the field in a homogeneous linear medium. In these cases, the modal decomposition of the cavity of quantization permits the separation of the overall Hamiltonian into a sum of mutually commuting partial Hamiltonians, one for each mode, each of which has the structure of a harmonic oscillator. In the presence of a nonlinear polarization, however, the modal approach does not produce such a simplification since the nonlinear interaction terms in the Hamiltonian cause the modes to mix with each other. This reflects the fact that reciprocal-space (i.e., Fourier-transform) techniques are helpful for solving linear differential equations by converting them into algebraic equations. For nonlinear differential equations, on the other hand, a Fourier transformation does not simplify the solution.

There is a class of problems, nevertheless, for which the modal approach is very convenient for calculating nonlinear dynamics of the field: Inside a cavity, when the field excitations extend throughout the cavity, boundary conditions impose a stationary modal structure to the field, involving a small number of modes. The nonlinear interaction term in the Hamiltonian, in such a case, reduces to a very simple form when it is expressed in terms of modal operators. The modal approach has been extensively used in the development of the input-output formalism for nonlinear interactions in cavities [7–9].

In propagative problems, on the other hand, we examine, generally, the interactions undergone by a short pulse of light (much shorter than the cavity) as it progresses in space through the nonlinear medium. Such propagative problems are traditionally treated through the modal approach by setting up wave packets of a large number of modes which move in space as the relative phases of the modes evolve in time under the zeroth-order field Hamiltonian and undergo nonlinear interactions under the nonlinear polarization term. This procedure can describe many of the features of traveling-wave phenomena but, quite often, it mixes effects related to the spatial progression of a beam with the spectral manifestations of the optical nonlinearity. For example, for the case of traveling-wave parametric generation [10], a wave-vector mismatch may appear as an energy (frequency) nonconservation term. In addition, in the presence of a strong nonlinearity and high light intensities, a modal analysis becomes extremely cumbersome as, for example, in the case of an intense light beam propagating in a medium with a strong nonlinear refractive index: The beam may undergo catastrophic self-focusing down to a point, a situation that involves a very large number of modes, all interacting with each other. The complexity of the modal approach in this problem contrasts with the simplicity of the standard viewpoint of classical nonlinear optics in which self-focusing is described relatively easily (albeit through a numerical solution) in terms of a spatial differential equation involving the local value of the electric field.

To circumvent the difficulties associated with the modal description of propagation, several authors have tried to recast the quantum-mechanical problem of propagation in direct space, so that it gives spatial differential equations analogous to those of classical nonlinear optics. One technique [6] involves the partition of the cavity of quantization into finite cells, in each of which we can define local field operators as the appropriate superpositions of the modal operators of the overall cavity. Another technique consists of considering the electromagnetic wave equation on the temporal Fourier components of the local electric-field operators [11,12]. As in classical nonlinear optics, in both cases, spatial differential equations are then obtained which may be solved to give the spatial structure of the electric-field operators.

In this paper, we present an alternate approach to the treatment of propagative phenomena in quantum optics. This approach is based directly on the canonical quantization procedure for the electromagnetic field in a homogeneous effective nonlinear medium, whereby the local field operators are quantized in direct space, with no reference to the normal modes of the cavity of quantization. Within this formalism, the spatial progression of the quantized operators is described by use of the momentum operator of the field, in addition to the Hamiltonian which describes its temporal evolution. The spatial and temporal coordinates of the field are thus treated on the same footing and can both be addressed through the algebraic techniques of second quantization. Some preliminary ideas have been given in two earlier publications [13,14], in which the problem of propagation through a linear medium was examined, however without following the canonical quantization procedure.

The paper is organized as follows: In Sec. II, we review the canonical quantization procedure for the electromagnetic field in a homogeneous, dispersionless, nonlinear medium. In Sec. III we discuss the description of propagative optical phenomena within the framework of a direct-space formulation of quantum optics, and we derive the operatorial equivalent of the Maxwell equations and the electromagnetic wave equation. In Sec. IV we illustrate the direct-space description of propagation (that is, without resorting to a modal decomposition of a propagating pulse) by examining the propagation of light through a linear medium and through a vacuum-dielectric interface. In Sec. V, we derive the operatorial equivalent of the slowly-varying-amplitude approximation, on which is based the classical theory of nonlinear optics, and in Sec. VI we apply this equation to the quantum-mechanical treatment of light propagation in a nonlinear medium. As an illustration of this quantum treatment, in Sec. VII, we examine the traveling-wave generation of squeezed light, by the degenerate down-conversion of a short pulse. Finally, in Sec. VIII, we summarize our conclusions.

II. FIELD QUANTIZATION IN AN EFFECTIVE MEDIUM

We consider the propagation of light in a lossless, nonmagnetic, homogeneous dielectric medium. We examine a simple geometry for the electromagnetic field such that the electric field E is polarized along the x axis, the magnetic field B along the y axis, while propagation occurs along the z axis. This assumption permits us to reduce all vectors to scalars, while at the same time it is representative of simple propagative experiments in which x-polarized light emerges from a source, traverses different (linear or nonlinear) optical elements placed along z, and then is detected. In this simple geometry the Maxwell equations reduce to two scalar differential equations,

$$\frac{\partial E}{\partial z}=-\frac{\partial B}{\partial t}\ , \tag{2.1a}$$

$$\frac{\partial B}{\partial z}=-\frac{\partial D}{\partial t}\ , \tag{2.1b}$$

where the displacement field D is defined by

$$D=E+P\ , \tag{2.2a}$$

with P being the polarization of the medium, which can be expressed as a converging power series of the electric field

$$P=\chi^{(1)}E+\chi^{(2)}E^2+\cdots+\chi^{(n)}E^n+\cdots\ , \tag{2.2b}$$

where $\chi^{(n)}$ is the nth-order optical susceptibility of the medium. We ignore its tensorial properties and its frequency dependence (dispersion). Dispersion cannot be taken into account rigorously within a quantum-mechanical theory based on the Hamiltonian formulation [5]. It is thus often introduced phenomenologically once a Hamiltonian has been obtained [6], since it may be important for nonlinear propagation. We use Heaviside-Lorentz units and take $\hbar=c=1$.

The Lagrangian density that incorporates the phenomenological definition of the polarization (2.2) and describes the field dynamics involved in the macroscopic Maxwell equations was shown [5,6] to be

$$\mathcal{L}=\frac{E^2-B^2}{2}+\tfrac{1}{2}\chi^{(1)}E^2+\tfrac{1}{3}\chi^{(2)}E^3+\tfrac{1}{4}\chi^{(3)}E^4+\cdots\ . \tag{2.3}$$

This effective Lagrangian density is the most general involving only the electric field (the medium is nonmagnetic) and possessing gauge invariance. One possible way of obtaining Eq. (2.3) rigorously from the standard Lagrangian of the electromagnetic field interacting with point charges, could involve first the use of the Power-Zienau-Woolley transformation [1,15] on the standard Lagrangian in order to incorporate the microscopic charge distribution into a macroscopic polarization density. Then, the atomic degrees of freedom could be eliminated.

We introduce the vector potential A and adopt the Coulomb gauge in which the scalar potential is taken as $\phi=0$ and A is transverse. In our simple geometry, the vector potential is related to the electric and magnetic fields by

$$E=-\frac{\partial A}{\partial t} \tag{2.4a}$$

and

$$B=\frac{\partial A}{\partial z}\ , \tag{2.4b}$$

with A polarized along the x axis. In a rigorous quantization procedure, the full vectorial representation of A (and also of E and B) is necessary, since the vacuum fluctuations are present on all components of A. In our simplified geometry, however, in which the propagation geometry is one dimensional and the susceptibilities are taken as scalars, the vectorial components of A are not coupled to each other and each component can be examined independently of the others. Thus our discussion may be limited only to that component of A which carries the excitation of the field, and a scalar notation may be used.

Note that the effective Lagrangian (2.3) is valid only at energies that are low with respect to those of the atomic excitations or atomic ionization. In fact, Eq. (2.3) leads to a theory that breaks down at high energies, since the nonlinear interaction terms give rise to incurable infinities in Feynman diagrams containing photon loops: the effective theory is not renormalizable, even in our simple one-dimensional propagation geometry because the nonlinear terms contain powers of the time derivative of A rather than of A itself. The correct interpretation of the effective theory, therefore, requires that no loops appear in a perturbative treatment based on Eq. (2.3) (tree approximation). The interaction coefficients $\chi^{(n)}$ of the effective theory account already for the loop diagrams that can be written in the microscopic theory, in which infinities *can* be eliminated (i.e., the microscopic theory is renormalizable).

As in the Power-Zienau-Woolley theory, the conjugate momentum of A with respect to the Lagrangian density (2.3) is the electric displacement

$$\Pi=\frac{\partial\mathcal{L}}{\partial\dot{A}}=-D\ . \tag{2.5}$$

Using this Lagrangian density, we may calculate the energy-momentum tensor [16] of the electromagnetic field inside a nonlinear medium $\Theta_{\mu\nu}$. Two elements of $\Theta_{\mu\nu}$ are of particular interest in our simple one-dimensional geometry, namely, the energy density,

$$\Theta_{tt}=\Pi\frac{\partial A}{\partial t}-\mathcal{L}=\frac{B^2+E^2}{2}+\tfrac{1}{2}\chi^{(1)}E^2+\tfrac{2}{3}\chi^{(2)}E^3+\tfrac{3}{4}\chi^{(3)}E^4+\cdots \tag{2.6}$$

and the momentum density,

$$\Theta_{tz}=-\Pi\frac{\partial A}{\partial z}=DB\ . \tag{2.7}$$

Each of these two elements, when integrated over the volume that contains the field (i.e., over infinite space), is independent of time and thus constitutes a constant of the motion. The spatially integrated quantities correspond, respectively, to the Hamiltonian and momentum operators that describe the temporal evolution and spatial progression of the electromagnetic field.

In setting up the Hamiltonian, the electric field E has to be expressed in terms of the electric displacement which is the canonical momentum of A according to Eq. (2.5). That is,

$$E=\beta^{(1)}D+\beta^{(2)}D^2+\beta^{(3)}D^3+\cdots , \tag{2.8}$$

where the β coefficients may be expressed in terms of the susceptibilities $\chi^{(n)}$ through the definition and Eq. (2.2), as [5]

$$\beta^{(1)}=\frac{1}{1+\chi^{(1)}}=\frac{1}{\epsilon} , \tag{2.9a}$$

$$\beta^{(2)}=-\beta^{(1)}\beta^{(1)}\beta^{(1)}\chi^{(2)} , \tag{2.9b}$$

and so on. The Hamiltonian is thus written as

$$H=\int_V \Theta_{tt}d\mathbf{r}=\int_V \tfrac{1}{2}B^2+\tfrac{1}{2}\beta^{(1)}D^2+\tfrac{1}{3}\beta^{(2)}D^3 + \tfrac{1}{4}\beta^{(3)}D^4+\cdots d\mathbf{r} , \tag{2.10}$$

while the momentum operator can be calculated as

$$G=\int_V \Theta_{tz}d\mathbf{r}=\int_V BD\,d\mathbf{r} , \tag{2.11}$$

where the integration is over the infinite cavity of quantization, obeying periodic boundary conditions, that is, $B(+\infty)=B(-\infty)$ and $D(+\infty)=D(-\infty)$.

We can now quantize the field by replacing each field variable by the corresponding operator and defining the equal-time commutator between the displacement D and vector potential A operators as [5,6]

$$[D(\mathbf{r},t),A(\mathbf{r}',t)]=i\delta_T(\mathbf{r}-\mathbf{r}') , \tag{2.12}$$

where in our simple geometry the transverse δ function δ_T reduces to the ordinary δ function. A further simplification of the canonical commutation relation (2.12) can be invoked in this paper, since we examine one-dimensional propagation along the z direction, with no transverse effects: we may consider that there is always an implicit integration over the x and y directions, so that the three-dimensional position vector $\mathbf{r}$ can be replaced by the coordinate z.

The vector potential A does not appear explicitly in the Hamiltonian (2.10) and momentum (2.11) operators, but rather in terms of its spatial derivative B. In view of this feature, the canonical commutator (2.12) may be expressed also in a form that is better suited to the use of the magnetic-field operator. Using the relationship between the magnetic field and the vector potential (2.4b), the canonical commutator may be rewritten in terms of the magnetic field as

$$[D(z,t),B(z',t)]=-i\delta'(z-z') , \tag{2.13}$$

where

$$\delta'(z-z')=\frac{d}{dz}\delta(z-z') \tag{2.14}$$

is the derivative of the δ function.

At this point, there are two equivalent representations within which the theory of quantum optics may be developed: reciprocal or direct space. The conventional theory of quantum optics adopts the reciprocal-space representation by expanding the field operators in terms of normal modes, thus defining modal (photon) creation and annihilation operators. The reciprocal-space representation is more appropriate for an eigenstate analysis of the field. In this paper, on the other hand, we shall develop a direct-space approach to quantum optics which avoids the modal decomposition of the field and uses local operators for the field observables. All calculations involving these local operators can be carried out by use of the direct-space commutator (2.12) on which is based the quantization of the field. This approach is more convenient for examining the propagation of a short pulse of light through a linear or nonlinear medium.

One last point we shall discuss in this section is the question of operator ordering. Because of the introduction of the commutator (2.12), the definition of the momentum operator given by Eq. (2.11) differs by an infinite quantity from the momentum operator in which B and D are permuted. The same is true for any operator that consists of a product of A, D, E, or B operators. To avoid this ambiguity, we shall consider any product of noncommuting operators that appears in an expression as being fully symmetrized (i.e., including all possible permutations of the individual field operators), such as

$$BD^2 \Longrightarrow \frac{BD^2+DBD+D^2B}{3} . \tag{2.15}$$

III. PROPAGATION IN DIRECT-SPACE QUANTUM OPTICS

In classical optics, the problem of the propagation of light through direct space is often addressed by considering the spatial structure of a wave whose time dependence is specified beforehand, such as, for example, a monochromatic wave oscillating in time. For such a wave, the classical electromagnetic wave equation reduces to a spatial differential equation (such as the Helmholtz equation of linear optics) whose solution gives directly the spatial progression of the wave.

This relatively simple procedure, however, cannot be directly transposed to quantum optics because this latter theory, like every quantum-mechanical theory, is based on the Hamiltonian formulation of mechanics, in which the time variable plays a particular role. That is, the standard quantization procedure, based on the equal-time commutator (2.12), as outlined in Sec. II, requires that the field be specified over all space at one instant of time (e.g., at $t=0$). In fact, it is this requirement that permits integration of the energy and momentum densities over all space in Eqs. (2.10) and (2.11). All the subsequent evolution of the field may then be calculated by using the Hamiltonian (2.10) which contains all the dynamical information on the field inside the medium. Thus, within the Hamiltonian formulation of quantum optics, a treatment of propagative phenomena in which the time dependence of the field is specified beforehand (as in classical optics) requires the introduction of a source term in the

Hamiltonian, such that it generates a time-varying (e.g., oscillating) field. The propagating electromagnetic field would thus be treated as a driven system.

In this paper, we treat source-free propagation within the Hamiltonian formalism, specifying the propagating wave by its initial spatial distribution rather than by its temporal characteristics. For a propagating short pulse of light, this would correspond to a "snapshot" of its evolution at one instant of time, for example, when the pulse is located at the entrance of a nonlinear crystal. Propagation, then, consists of the subsequent time evolution of the initial distribution of the field, under the Hamiltonian relevant to the effective medium. Thus, this approach can rely directly on the Hamiltonian of Eq. (2.10).

The Hamiltonian of the electromagnetic field relates the spatial distribution of the field at t to its spatial distribution at another instant $t+dt$ later through the Heisenberg equation, which can be written as

$$\frac{\partial Q}{\partial t}=i[H,Q]=iH^{\times}Q \ , \tag{3.1}$$

where Q is any field operator. We shall use hereafter the Kubo notation [17] for the commutator, whereby the superscript $\times$ denotes the commutation of the operator it superscribes with everything that follows. One of the advantages of this compact notation is that it permits a simple formal solution of the Heisenberg equation (3.1), to give the time evolution of the Q operator as

$$Q(t)=e^{itH^{\times}}Q(0) \tag{3.2a}$$

as it can be verified by expanding the exponential,

$$\begin{aligned} e^{itH^{\times}}Q(0)&=Q(0)+(it)H^{\times}Q(0)\\ &\quad+\frac{1}{2!}(it)^2H^{\times}H^{\times}Q(0)+\cdots\\ &=Q(0)+(it)[H,Q(0)]\\ &\quad+\frac{1}{2!}(it)^2[H,[H,Q(0)]]+\cdots\\ &=e^{iHt}Q(0)e^{-iHt} \ . \end{aligned} \tag{3.2b}$$

Similarly, the momentum operator G relates the operator Q at a point on the z axis to another point at $z+dz$ (at the same instant of time) through the Heisenberg-like equation involving the momentum

$$\frac{\partial Q}{\partial z}=-iG^{\times}Q \ . \tag{3.3}$$

This equation can be solved formally to describe the translation of Q from the point z_0 to the point z in direct space as

$$Q(z)=e^{-i(z-z_0)G^{\times}}Q(z_0) \ , \tag{3.4}$$

in a manner analogous to the way that the Hamiltonian gives the temporal evolution of Q.

Within the conventional modal approach to quantum optics, the Heisenberg equation involving the Hamiltonian (3.1) is used alone in describing the dynamics of the field, while the Heisenberg equation involving the momentum operator (3.3) is ignored. The reason is that the modes are eigenstates of the momentum operator G, and this makes the explicit use of the differential equation (3.3) superfluous, since its solution gives simply the spatially dependent phase factor e^{ikz} associated with the delocalized modal operators. Within the direct-space approach, on the other hand, the momentum operator G is necessary to express the temporal evolution of the field in terms of its spatial progression: a direct-space field operator, such as $E(z,t)$, may be considered as a short pulse that corresponds to a field distribution around the point z at time t, and thus its time evolution involves a translation of the pulse along the z axis, a feature that is formalized in terms of the momentum operator G in Eq. (3.4).

The Hamiltonian (2.10) and momentum (2.11) operators can relate the electric- and magnetic-field operators to each other, through the direct-space commutator (2.12). As shown in Appendix A, this relationship is expressed by two commutator equations,

$$G^{\times}E=H^{\times}B \tag{3.5a}$$

and

$$G^{\times}B=H^{\times}D \ . \tag{3.5b}$$

These two equations may be considered as the operatorial equivalent of the Maxwell equations (2.1), since the H and G commutators are directly related to the t and z derivatives, according to the Heisenberg equations (3.1) and (3.3). Using the fact that, in a homogeneous medium, the Hamiltonian and momentum operators commute with each other, that is

$$G^{\times}H=0 \ , \tag{3.6}$$

Eqs. (3.5) may be combined into the operatorial equivalent of the electromagnetic wave equation,

$$G^{\times}G^{\times}E=H^{\times}H^{\times}D \ , \tag{3.7}$$

as is usually done for the classical Maxwell equations.

This operatorial wave equation is as general as the corresponding classical equation, and is at the root of the formalism developed in this paper for the direct-space description of light propagation: it provides a rule for relating multiple powers of the Hamiltonian commutator in the expansion of the time-evolution operator (3.2b) to multiple powers of the momentum commutator. Thus it permits a description of the temporal evolution of a short light pulse in terms of its progression in direct space, without requiring a modal analysis of the pulse. This approach for the description of propagation makes contact between the operatorial methods of quantum optics and the differential equations commonly used in classical optics. In this way, the results of classical propagative optics can readily be translated into an operatorial language and applied to the treatment of propagation in the quantized electromagnetic field.

IV. PROPAGATION IN A LINEAR MEDIUM

To illustrate the direct-space approach to quantum optics, we shall examine here the problem of propagation of a short light pulse in a linear (refractive) medium. To make contact with the well-known results of classical linear optics, we develop here the quantum-mechanical formalism in terms of the electric and magnetic fields E and B, rather than in terms of D and B which are the canonical variables. For a linear medium, the operatorial wave equation (3.7) reduces to

$$(G^{\times}G^{\times}-\epsilon H^{\times}H^{\times})E=0 , \tag{4.1}$$

where $\epsilon=1+\chi^{(1)}$ is the dielectric function of the medium. It is also convenient to define $v=1/\sqrt{\epsilon}$, the velocity of an electromagnetic wave in the refractive medium. For free space, the wave equation is identical to Eq. (4.1), however with $\epsilon=1$ and $v=c$. In this section, for the sake of clarity, we shall introduce explicitly c, the speed of light, in order to identify light waves propagating in free space.

The electromagnetic field is defined over all space at $t=0$ through the operators $E(z,0)$ and $B(z,0)$. At a later time t, the electric field $E(z,t)$ is related to the electric field at the same point in space z but at an earlier time ($t=0$) by

$$E(z,t)=e^{itH^{\times}}E(z,0) . \tag{4.2a}$$

This exponential can be expanded as

$$\begin{aligned} E(z,t)=&E(z,0)+(it)H^{\times}E(z,0)\\ &+\frac{1}{2!}(it)^2H^{\times}H^{\times}E(z,0)\\ &+\frac{1}{3!}(it)^3H^{\times}H^{\times}H^{\times}E(z,0)+\cdots . \end{aligned} \tag{4.2b}$$

The wave equation for a linear medium (4.1) permits us to replace all pairs of H commutators in the expansion of Eq. (4.2b) by an equal number of pairs of G commutators. Similarly, the Maxwell equation (3.5b) permits us to rewrite all odd powers of the expansion (4.2b) as G commutators of the initial distribution of the magnetic field $B(z,0)$. Equation (4.2) thus becomes

$$\begin{aligned} E(z,t)=&E(z,0)+\frac{1}{2!}(ivt)^2G^{\times}G^{\times}E(z,0)+\frac{1}{4!}(ivt)^4G^{\times}G^{\times}G^{\times}G^{\times}E(z,0)+\cdots\\ &+(ivt)vG^{\times}B(z,0)+\frac{1}{3!}(ivt)^3vG^{\times}G^{\times}G^{\times}B(z,0)+\cdots\\ =&\cos(vtG^{\times})E(z,0)+iv\sin(vtG^{\times})B(z,0) . \end{aligned} \tag{4.3}$$

Similarly for the magnetic field, the operatorial Maxwell equations can be combined into a wave equation of the form

$$(G^{\times}G^{\times}-\epsilon H^{\times}H^{\times})B=0 , \tag{4.4}$$

which gives

$$B(z,t)=\cos(vtG^{\times})B(z,0)+\frac{i}{v}\sin(vtG^{\times})E(z,0) . \tag{4.5}$$

Equations (4.3) and (4.5) indicate that the linear combination

$$W_v^+(z,t)=E(z,t)+vB(z,t) \tag{4.6}$$

evolves in time as

$$W_v^+(z,t)=e^{ivtG^{\times}}W_v^+(z,0)=W_v^+(z-vt,0) . \tag{4.7}$$

That is, the operator W_v^+ represents an electromagnetic wave moving towards $+\infty$ at a constant speed of $v=c/\sqrt{\epsilon}$, retaining its original form. Similarly, the linear combination

$$W_v^-(z,t)=E(z,t)-vB(z,t) \tag{4.8}$$

evolves as

$$W_v^-(z,t)=e^{-ivtG^{\times}}W_v^-(z,0)=W_v^-(z+vt,0) \tag{4.9}$$

and corresponds to a backward-moving wave (towards $-\infty$) at the same speed. We note that, in this direct-space description for the propagation of a short light pulse inside a linear medium, no modal decomposition of the field was necessary.

The problem of a vacuum-dielectric interface is addressed in a way very similar to that of classical optics, in terms of the boundary conditions. To examine the problem of the interface, we now consider two half-spaces, such that the $z=(-\infty,0)$ half-space is empty (i.e., $v=c$) while the $z=(0,+\infty)$ half-space consists of a transparent linear dielectric (with $v<c$). We also consider a short pulse which, at $t=0$, is contained completely in the empty half-space and has the form $W_c^+(z_0,0)$ with $z_0<0$. As time evolves, this pulse propagates towards the interface. Beyond the interface, in the refractive medium, the pulse must assume the form W_v^+ with $v<c$. Continuity of the electric and magnetic fields at the interface requires, therefore, that a backward-going wave of the form W_c^- must also be generated in the empty half-space. Matching the amplitudes of the three waves across the interface, we may obtain, as in classical optics, the transmission and reflection coefficients for the electric-field operator as

$$t=\frac{2\sqrt{\epsilon}}{\sqrt{\epsilon}+1}, \quad r=-\frac{\sqrt{\epsilon}-1}{\sqrt{\epsilon}+1} . \tag{4.10}$$

It is relatively straightforward now to verify that the equal-time commutator of the electric- and magnetic-field operators is preserved at all times, if we consider the transmitted and reflected waves. To this end, we consider two very short electromagnetic pulses in the empty half-space, $W_c^+(z_0,0)$ and $W_c^+(z_0',0)$ described as distributions of the variables z_0 and z_0' ($z_0,z_0'<0$) at $t=0$. In the empty half-space, at $t=0$, the electric and magnetic fields satisfy the equal-time commutator

$$[E(z_0,0)]^{\times}B(z_0',0)=-i\delta'(z_0-z_0') \ . \tag{4.11}$$

At time t ($t>|z_0|/c$), the two pulses are partly transmitted into the dielectric and partly reflected back into the empty half-space, so that each pulse can be written as a superposition of the two beams into which it is split. That is, $W_c^+(z_0,0)$ evolves into

$$\frac{2\sqrt{\epsilon}}{\sqrt{\epsilon}+1}W_v^+(\zeta,t)-\frac{\sqrt{\epsilon}-1}{\sqrt{\epsilon}+1}W_c^-(z,t) \ , \tag{4.12a}$$

where

$$z=-c\left[t-\frac{|z_0|}{c}\right] \tag{4.12b}$$

is located in the empty half-space, while

$$\zeta=v\left[t-\frac{|z_0|}{c}\right] \tag{4.12c}$$

is in the dielectric. A similar expression also holds for the evolution of $W_c^+(z_0',0)$.

Using the relationship between z and z_0 imposed by propagation [Eq. (4.12b)] (and also between z' and z_0') and the commutator (3.10) applied to free space (i.e., with $D=E$), we may calculate the equal-time commutator for the electric and magnetic fields of the reflected waves as

$$[E(z,t)]^{\times}B(z',t)=-i\delta'(z-z')=-i\delta'(z_0-z_0') \ . \tag{4.13}$$

For the transmitted waves we have to use the equal-time commutation relation between the displacement and the magnetic fields *inside the dielectric* which is

$$[\epsilon E(\zeta,t)]^{\times}B(\zeta',t)=-i\delta'(\zeta-\zeta') \ . \tag{4.14}$$

Again, introducing the relationship between ζ and z_0 (and ζ' and z_0') (4.12c), together with the property of the derivative of the δ function

$$\delta'(\alpha x)=\frac{1}{\alpha^2}\delta'(x) \tag{4.15}$$

the commutator (4.14) becomes

$$[E(\zeta,t)]^{\times}B(\zeta',t)=-i\delta'(z_0-z_0') \ . \tag{4.16}$$

Thus, by using the commutators for the reflected (4.13) and refracted (4.16) waves, together with the expression for the electromagnetic wave after beamsplitting (4.12), we may calculate the overall equal-time commutator after beamsplitting as

$$\begin{aligned}[E(t)]^{\times}B(t)&=\frac{4\sqrt{\epsilon}}{(\sqrt{\epsilon}+1)^2}\{[E(\zeta,t)]^{\times}B(\zeta',t)\}\\&\quad+\left[\frac{\sqrt{\epsilon}-1}{\sqrt{\epsilon}+1}\right]^2\{[E(z,t)]^{\times}B(z',t)\}\\&=-i\delta'(z_0-z_0') \ ,\end{aligned} \tag{4.17}$$

which is identical to the commutator before beamsplitting (4.11). Thus, consideration of the three waves, incident, reflected, and transmitted, each evolving in time under the Hamiltonian relevant to its medium, leads to the preservation of the equal-time commutator of the electromagnetic field for all time.

V. THE OPERATORIAL SLOWLY-VARYING-AMPLITUDE WAVE EQUATION

As in classical optics, the problem of propagation of a short pulse in a nonlinear medium cannot be solved in the general case. In most cases of interest, however, the energy associated with the nonlinear part of the Hamiltonian (2.4) is much smaller than the energy of the linear part. This implies that the effect of the nonlinear polarization on the time evolution of an electromagnetic wave can be treated as a perturbation with respect to the variation of the wave under the linear Hamiltonian and momentum operators. In classical nonlinear optics, these considerations give rise to the slowly-varying-amplitude (SVA) approximation [18] of the electromagnetic wave equation. In this section we shall examine a perturbative treatment of the time evolution of the field in a nonlinear medium, that corresponds to propagation within the SVA approximation. To simplify the discussion we consider a nonlinear medium that displays only one order of nonlinearity, characterized by a single nonlinear susceptibility $\chi^{(n)}$.

In order to develop a perturbative treatment of nonlinear propagation, we consider that the optical nonlinearity of the medium is absent at $t=-\infty$ and is turned-on adiabatically. At $t=-\infty$, in the absence of the nonlinearity, the electric and magnetic fields in the medium, E_0 and B_0, as well as the displacement field which is given by

$$D_0=\epsilon E_0 \ , \tag{5.1}$$

propagate under the Hamiltonian

$$H_0=\int\frac{B_0^2+D_0^2/\epsilon}{2}d\mathbf{r} \tag{5.2a}$$

and momentum operators

$$G_0=\int B_0D_0d\mathbf{r} \tag{5.2b}$$

which include only the effects of the linear polarization of the medium and are, therefore, of zeroth order in the nonlinear susceptibility $\chi^{(n)}$. Thus D_0 and B_0 vary in scales of the order of the optical period and optical wavelength and are the fields that would have existed in the medium at finite t, if the nonlinearity were absent. We note that these fields correspond to the traditional "in" fields of scattering theory, and can thus serve as a basis for the perturbative treatment of the exact fields D and B, interacting through the nonlinear susceptibility.

After the nonlinearity is "on," the zeroth-order Hamiltonian turns into the exact Hamiltonian (2.10) and the displacement field D becomes a nonlinear function of the exact electric field E given by Eqs. (2.2) or (2.8). Following the standard perturbation theory [16], the exact field operators in the nonlinear medium can be related to the zeroth-order fields by the unitary transformation

$$D(z,t)=U^{-1}(t)D_0(z,t)U(t) \tag{5.3a}$$

and

$$B(z,t)=U^{-1}(t)B_0(z,t)U(t) \ . \tag{5.3b}$$

The unitarity of U ensures that the commutation relation (2.12) or (2.13) is preserved when the exact field operators are expressed in terms of zeroth-order operators. The transformation U is given by the time-ordered exponential

$$U(t)=T\exp\left[-i\lambda\int_{-\infty}^{t}\tilde{H}_1(\tau)d\tau\right] , \tag{5.4}$$

where

$$H_1=\frac{1}{n+1}\int\beta^{(n)}D_0^{n+1}d\mathbf{r}=-\frac{1}{n+1}\int\chi^{(n)}E_0^{n+1}d\mathbf{r} \tag{5.5}$$

is the nonlinear interaction part of the Hamiltonian. We note that, in Eq. (5.5), H_1 is written in terms of the zeroth-order fields D_0 and B_0, rather than in terms of the full interacting fields D and B, as it was in Eq. (2.10). Thus H_1 is first order in the nonlinear susceptibility $\chi^{(n)}$ (which give the energy scale associated with the nonlinear polarization) and the time-ordered exponential represents a perturbative expansion in successive powers of $\chi^{(n)}$. The dimensionless parameter λ has been introduced in Eq. (5.4) for the bookkeeping of the successive orders of the nonlinear coefficient in the perturbative expansion. The integrand of Eq. (5.4)

$$\tilde{H}_1(\tau)=e^{i\tau H_0^{\times}}H_1 \tag{5.6}$$

is the time evolution of the interaction Hamiltonian, under the linear Hamiltonian H_0.

Expanding the exponential (5.4), we can express the exact Hamiltonian (2.10) perturbatively, up to first order in λ as

$$\begin{aligned}H&=U^{-1}(t)[H_0+\lambda\tilde{H}_1(t)]U(t)\\&=H_0+\lambda\left[\tilde{H}_1(t)+i\int_{-\infty}^{t}d\tau[\tilde{H}_1(\tau)]^{\times}H_0\right]+O(\lambda^2) \ .\end{aligned} \tag{5.7}$$

To calculate the first-order term in Eq. (5.7), we partition the interaction Hamiltonian H_1 into two parts,

$$H_1=H_{1S}+H_{1N} \tag{5.8a}$$

such that H_{1S} commutes with the linear Hamiltonian H_0

$$H_{1S}^{\times}H_0=0 , \tag{5.8b}$$

while H_{1N} does not

$$H_{1N}^{\times}H_0\neq 0 \ . \tag{5.8c}$$

The form of H_{1S} and the choices that make it unique are discussed in Appendix B. We note that all three operators H_1, H_{1S}, and H_{1N} commute with the linear momentum operator G_0, as shown also in Appendix B. Using Eqs. (5.8), the integral in the first-order term in (5.7) can be written as

$$\begin{aligned}&i\int_{-\infty}^{t}d\tau[\tilde{H}_{1S}(\tau)]^{\times}H_0+i\int_{-\infty}^{t}d\tau[\tilde{H}_{1N}(\tau)]^{\times}H_0\\&\qquad=-\int_{-\infty}^{t}\frac{d}{d\tau}\tilde{H}_{1N}(\tau)d\tau=-\tilde{H}_{1N}(t) \ .\end{aligned} \tag{5.9}$$

Thus the nonlinear Hamiltonian can be written in terms of the linear-medium operators, up to first order in λ as

$$H=H_0+\lambda H_{1S}+O(\lambda^2) \ . \tag{5.10}$$

Equation (5.10) is simply a generalization of the well-known feature of first-order perturbation theory, whereby the first-order correction to the energy consists of the diagonal elements of the perturbation. Here, rather than consider as the "diagonal" part of H_1 only the part that leaves the state of the field unchanged after it operates, we include also the interactions that induce transitions which change the zeroth-order state of the field without however changing the (zeroth-order) energy.

The partition (5.8) may be easily related to more familiar concepts, if we express the interaction Hamiltonian H_1 in terms of the modes of H_0. In such a modal representation, the stationary part H_{1S} corresponds to the rotating-wave-approximation (RWA) terms of the nonlinear interaction. That is, for an nth-order nonlinearity, H_{1S} consists of all the $n+1$-fold products of photon creation and annihilation operators which conserve energy and describe all the resonant interactions among the modes. For example, for a quadratic nonlinearity, as can be verified by writing Eq. (B12a) in terms of modal creation and annihilation operators, H_{1S} consists of the sum of all terms of the form

$$b_j^{\dagger}b_l^{\dagger}b_m+b_m^{\dagger}b_lb_j \tag{5.11a}$$

such that $\omega_j+\omega_l=\omega_m$ and $k_j+k_l=k_m$. $b_j^{\dagger}$ and b_j are the familiar creation and annihilation operators for the jth mode of H_0. The nonstationary part H_{1N}, on the other hand, includes all the non-RWA terms which correspond to photon conversions that do not conserve energy to zeroth order, and describe the nonresonant interactions among the modes. In particular, for a quadratic nonlinearity H_{1N} includes terms of the form

$$b_j^{\dagger}b_l^{\dagger}b_m^{\dagger}+b_mb_lb_j \ , \tag{5.11b}$$

with arbitrary frequency relations, as well as nonresonant terms of the form (5.11a) such that $\omega_j+\omega_l\neq\omega_m$.

Within the perturbative treatment outlined above, the time evolution of the displacement operator can be written as

$$D(z,t)=e^{itH^{\times}}D(z,0)$$
$$=e^{it[H_0^{\times}+\lambda H_{1S}^{\times}+O(\lambda^2)]}$$
$$\times[D_0(z,0)+\lambda D_1(z,0)+O(\lambda^2)]\ , \qquad (5.12)$$

where

$$D_1(z,t)=i\int_{-\infty}^{t}d\tau[\tilde{H}_1(\tau)]^{\times}D_0(z,t) \qquad (5.13)$$

is the first-order correction to the displacement field. A similar expression also holds for the magnetic-field operator. In principle, within a strict first-order perturbative treatment, the exponential time-evolution operator in Eq. (5.12) should be expanded and only the first-order term retained. However, the amplitude of the first-order term is proportional to t and, therefore, exhibits secular behavior; that is, it grows without bounds at long times. Thus, in long time scales (i.e., in time scales much longer than the optical period and long enough that the effect of the optical nonlinearity is measurable macroscopically) it is a better approximation to the overall time evolution to keep all powers of H_{1S}, so that the amplitude of D can remain bounded even at very long times. In other words, from each order of the perturbative expansion of the exponential, only the term which is given by the nth power of the first-order correction to the Hamiltonian is retained. Within this approximation, Eq. (5.12) can be written as

$$D(z,t)\approx[e^{itH_{1S}^{\times}}D_0(z,t)]+D_1(z,t)\ , \qquad (5.14)$$

where D_0 and D_1 evolve under the linear-medium Hamiltonian H_0, and thus vary in scales of the order of the optical period and the optical wavelength. The approximation (5.14) in which the *exponent* of the time-evolution operator is treated perturbatively to first order is equivalent to making a first-order approximation on the *temporal derivative* of D, and is thus suitable for a perturbative treatment of the wave equation (3.7) which involves such derivatives.

The first-order approximation to the time-evolution operator discussed above can also be viewed in a way that makes contact with the language of classical nonlinear optics. The time-evolution operator $e^{iH_{1S}t}$ operating on D_0 in Eq. (5.14) may be regarded as a slowly varying envelope function imprinted by the nonlinear medium on the fast varying ("carrier") wave D_0 in the course of its propagation through the medium. This slow temporal modulation gives rise to a modification of the spatial progression of the wave, manifested on a long spatial scale, much longer than the optical wavelength and of the order of the propagation length over which the nonlinear effect becomes macroscopically measurable.

The relation between the slow temporal modulation of a wave propagating in a nonlinear medium and its long-scale spatial variation can be obtained from the nonlinear wave equation (3.7). By use of the unitary transformation (5.3) and its perturbative expansion (5.4), the exact nonlinear wave equation (3.7) can be written up to first order in λ as

$$G_0^{\times}G_0^{\times}\left[\frac{D_0+\lambda D_1-\lambda P_{\mathrm{NL}}(D_0)}{\epsilon}\right]$$
$$=(H_0^{\times}H_0^{\times}+2\lambda H_0^{\times}H_{1S}^{\times})(D_0+\lambda D_1)\ , \qquad (5.15)$$

where

$$P_{\mathrm{NL}}=-\epsilon\beta^{(n)}D_0^n=\chi^{(n)}E_0^n \qquad (5.16)$$

is the nonlinear polarization. Collecting terms that are of the same order in λ we can separate the exact nonlinear wave equation into a perturbative hierarchy of partial wave equations, one for each order of λ.

To order λ^0, the nonlinear wave equation gives

$$(G_0^{\times}G_0^{\times}-\epsilon H_0^{\times}H_0^{\times})D_0=0 \qquad (5.17)$$

identical to the linear wave equation that describes the spatial progression of D_0 as it evolves under the linear Hamiltonian H_0. Thus D_0 can be expressed as the sum of a forward- and a backward-going electromagnetic wave, as discussed in Sec. IV.

To order λ^1 we have

$$2\epsilon H_0^{\times}H_{1S}^{\times}D_0=-\epsilon H_0^{\times}H_0^{\times}D_1+G_0^{\times}G_0^{\times}D_1-G_0^{\times}G_0^{\times}P_{\mathrm{NL}}\ . \qquad (5.18)$$

The first-order wave equation can be put in a form that makes contact with the classical SVA wave equation of nonlinear optics, by introducing the definition (5.13) of D_1. However, rather than calculate explicitly the form of D_1, we can deduce its contribution to the wave equation (5.18) through a simple argument: Since D_0 obeys the zeroth-order wave equation (5.17), and since H_{1S} commutes both with H_0 and with G_0, the term in the left-hand side of Eq. (5.18) also obeys that zeroth-order wave equation. This means that the sum of all the terms on the right-hand side of Eq. (5.18) must also obey the zeroth-order wave equation. We now separate P_{NL} into two parts, P_W and $(P_{\mathrm{NL}}-P_W)$, such that P_W obeys the zeroth-order wave equation

$$(G_0^{\times}G_0^{\times}-\epsilon H_0^{\times}H_0^{\times})P_W=0\ . \qquad (5.19)$$

As discussed in Appendix B, this partition corresponds to the elimination of all terms that couple opposite-going waves in P_{NL}. Clearly, then, the two terms that involve D_1 in Eq. (5.18) serve to cancel $(P_{\mathrm{NL}}-P_W)$, which contains all the terms that consist of products of opposite-going waves.

In view of the above discussion, then, the first-order wave equation (5.18) can be written as

$$2\epsilon H_0^{\times}H_{1S}^{\times}D_0=-G_0^{\times}G_0^{\times}P_W\ . \qquad (5.20)$$

This equation relates the variation of the slowly varying envelope of a propagating wave (expressed by H_{1S}) to the nonlinear driving term. It is thus the operatorial equivalent of the classical SVA wave equation, which is usually written as

$$2ik\frac{\partial}{\partial z}\tilde{E}=\frac{\partial^2}{\partial t^2}P_W \qquad (5.21a)$$

or, more often, in terms of the individual temporal Fourier components of $\tilde{E}$ and P_W as

$$\frac{\partial}{\partial z}\tilde{E}(\omega)=\frac{i\omega}{2\sqrt{\epsilon}}P_W(\omega) , \tag{5.21b}$$

where $\tilde{E}$ is the envelope function of the electric field. We note that in Eqs. (5.20) and (5.21) the roles of the temporal and spatial derivatives are interchanged. Nevertheless, the two equations are equivalent, as can be seen by comparing their right-hand sides: Since P_W obeys the zeroth-order wave equation (5.19), its second spatial derivative [in Eq. (5.20)] is proportional to its second temporal derivative [in Eq. (5.21)]. The equivalence of the left-hand sides, that is the relation of the temporal evolution of the envelope function (characterized by H_{1S}) to its spatial progression, is discussed in Sec. VI.

Before closing this section we examine briefly the structure of P_W. A more complete discussion, as well as all the relevant calculations, appear in Appendix B. Equation (5.19) implies that P_W propagates in the same way as a simple electromagnetic wave in the linear medium, and consists of the "phase-matched" part of the nonlinear polarization. For example, for time-harmonic plane waves P_W corresponds to a truncation of $P_{\rm NL}$ in which are retained only those terms of the nonlinear polarization that oscillate at the same frequency and have the same wave vector as the propagating electromagnetic wave that is driven by this polarization, and thus vary as $\exp[i\omega(t-z\sqrt{\epsilon}/c)]$. For a quadratic nonlinearity, P_W involves a sum of terms of the form

$$b_j^\dagger b_l^\dagger+b_l b_j, \quad b_m^\dagger b_n , \tag{5.22}$$

such that $k_j+k_l=(\omega_j+\omega_l)\sqrt{\epsilon}/c$ and $k_m-k_n=(\omega_m-\omega_n)\sqrt{\epsilon}/c$, as can be verified by expressing Eq. (B10a) in terms of modal creation and annihilation operators.

VI. PROPAGATION IN A NONLINEAR MEDIUM

Equations (5.17) and (5.20) are the two wave equations that govern the propagation of the displacement field D in the two temporal and spatial scales in which its variations occur. The linear wave equation (5.17) describes the fast component (carrier wave) of the propagation of D, while the quantum-mechanical SVA equation (5.20) should permit a description of the spatial progression of the modulation envelope which the nonlinear medium imprints on the carrier wave.

We note, however, that while the zeroth-order wave equation (5.17) provides a simple substitutional rule for converting the time-evolution operator e^{iH_0t} into a spatial progression operator e^{iG_0z} (as discussed in Sec. IV), the form of the quantum-mechanical SVA equation is not as simple. Indeed, it presents two features that prevent such a straightforward conversion of the slow temporal modulation operator $e^{iH_{1S}t}$ into a SVA spatial envelope. First, the right-hand side of Eq. (5.20) is written in terms of the spatial derivative of the nonlinear polarization P_W, rather than in terms of the D field, as in the left-hand side. Second, the slow modulation expressed by H_{1S} on the left-hand side, is "entangled" with the fast component of propagation corresponding to H_0.

The first problem can be remedied by reexpressing the spatial dependence of the driving term P_W as a spatially dependent interaction of D_0 with itself. This can be done by defining an effective SVA "momentum" operator such that it obeys

$$G_{\rm SVA}^\times D_0=\tfrac{1}{2}G_0^\times P_W . \tag{6.1}$$

In this definition $G_{\rm SVA}$ expresses the modification of the spatial progression of D, because of the nonlinear interaction that it undergoes in the course of its propagation. The structure of $G_{\rm SVA}$ is examined in detail in Appendix B. Here we note simply that if we define an effective "interaction" momentum operator as

$$G_1=\tfrac{1}{2}\int B_0 P_{\rm NL}\,d\mathbf{r} \tag{6.2}$$

then this operator satisfies

$$G_1^\times D_0=\tfrac{1}{2}G_0^\times P_{\rm NL} \tag{6.3}$$

and thus $G_{\rm SVA}$ can be considered as the RWA part of G_1, such that it commutes with H_0. The effective SVA momentum operator, thus, can be considered as the spatial equivalent of the stationary part of the interaction Hamiltonian H_{1S} and has a similar structure. Indeed, for the case of a quadratic nonlinearity, $G_{\rm SVA}$ consists of the sum of all terms of the form

$$b_j^\dagger b_l^\dagger b_m+b_m^\dagger b_l b_j \tag{6.4}$$

such that $\omega_j+\omega_l=\omega_m$ and $k_j+k_l=k_m$, analogous to the terms (5.11a), however with different coefficients from those of H_{1S}.

With the definition (6.1), the SVA wave equation (5.20) can be written as

$$(G_{\rm SVA}^\times G_0^\times+\epsilon H_{1S}^\times H_0^\times)D_0=0 . \tag{6.5}$$

In this form, the operatorial SVA equation relates directly the slow component of the temporal evolution of a short pulse of the displacement field D_0, to its spatial progression: $G_{\rm SVA}$ describes the long-scale spatially dependent distortion of the pulse.

We now come to the problem of the "entanglement" of the fast and slow components of propagation, which is expressed in Eq. (6.5) by the fact that H_{1S} and $G_{\rm SVA}$ occur each in a product with H_0 and G_0, respectively. This arises because the electric-field wave (represented by D_0) alternates between the electric and magnetic fields in the course of its propagation under the linear Hamiltonian, as seen in Eq. (4.3). Thus, in order to disentangle the fast and slow variations, it is necessary to consider at the same time the SVA equation for the magnetic field, as in Sec. IV. To this end, define forward (+) and backward (−) polarization waves in the nonlinear medium as

$$V^{\pm}=D\pm\sqrt{\epsilon}B , \tag{6.6}$$

so that in the absence of the nonlinearity these waves reduce to the corresponding electromagnetic waves in the linear medium

$$V_0^{\pm} = D_0 \pm \sqrt{\epsilon} B_0 = \epsilon W_v^{\pm} , \tag{6.7a}$$

with W_v^+ and W_v^- defined in a way analogous to Eqs. (4.6) and (4.8), in terms of E_0 and B_0; that is,

$$W_v^{\pm} = E_0 \pm v B_0 . \tag{6.7b}$$

The factor $\sqrt{\epsilon}$ in Eq. (6.6) ensures that the effects of the linear polarization are treated exactly in the propagation of $V^{\pm}$, while the effects of the nonlinear polarization can be treated perturbatively. Using the first-order treatment developed in Sec. V, the temporal evolution of the V^+ can be written as

$$V^+(z,t) \approx [e^{itH_{1S}^{\times}} \epsilon W_v^+(z,t)] + V_1^+(z,t) , \tag{6.8}$$

where V_1^+ is the first-order correction to V given by an equation analogous to (5.13). The wave equation involving the polarization wave,

$$(G^{\times} - \sqrt{\epsilon} H^{\times}) V^+ = -G^{\times} P_{\rm NL} , \tag{6.9}$$

can then be treated perturbatively to give a hierarchy of partial wave equations. Thus, to zeroth order in the optical nonlinearity, we have

$$(G_0^{\times} - \sqrt{\epsilon} H_0^{\times}) W_v^+ = 0 , \tag{6.10}$$

which is the equation for a forward-going electromagnetic wave in a linear medium, while to first order we have

$$\sqrt{\epsilon} H_{1S}^{\times} W_v^+ = -\sqrt{\epsilon} H_0^{\times} V_1^+ + G_0^{\times} V_1^+ - G_0^{\times} P_{\rm NL} . \tag{6.11}$$

As discussed in Eq. (5.20), the contribution of the D_1 part of V_1 reduces $P_{\rm NL}$ to P_W, so that the right-hand side of Eq. (6.11) can be written as

$$(G_0^{\times} - \sqrt{\epsilon} H_0^{\times}) \int_{-\infty}^{t} \tilde{H}_1^{\times} (D_0 + \sqrt{\epsilon} B_0) d\tau - G_0^{\times} P_{\rm NL}$$
$$= (G_0^{\times} - \sqrt{\epsilon} H_0^{\times}) \int_{-\infty}^{t} \tilde{H}_1^{\times} (\sqrt{\epsilon} B_0) d\tau - G_0^{\times} P_W . \tag{6.12a}$$

Using the definition of the effective SVA momentum (6.1) and expressing D_0 in terms of W_v^+ and W_v^-, Eq. (6.12a) can be rewritten as

$$(G_0^{\times} - \sqrt{\epsilon} H_0^{\times}) \int_{-\infty}^{t} \tilde{H}_1^{\times} (\sqrt{\epsilon} B_0) d\tau - G_{\rm SVA}^{\times} (W_v^+ + W_v^-) . \tag{6.12b}$$

We note that the left-hand side of the first-order wave equation (6.11) is a solution of the zeroth-order forward-wave equation (6.10), which implies that the right-hand side of Eq. (6.11) must also satisfy that zeroth-order forward-wave equation. Thus the first-order correction to the magnetic-field operator in Eq. (6.12b) serves essentially to cancel the contribution of the backward-going wave W_v^-, so that the first-order wave equation (6.11) reduces to

$$\sqrt{\epsilon} H_{1S}^{\times} W_v^+ = -G_{\rm SVA}^{\times} W_v^+ . \tag{6.13a}$$

Similarly, for the backward-going wave we have

$$\sqrt{\epsilon} H_{1S}^{\times} W_v^- = G_{\rm SVA}^{\times} W_v^- . \tag{6.13b}$$

Equations (6.13) provide a simple rule for substituting H_{1S} by $vG_{\rm SVA}$ to convert the temporal evolution of the modulation envelope into a spatial progression. That is, the overall propagation of a polarization wave in a nonlinear medium can be written within the SVA approximation as

$$V^+(z,t) = \epsilon[e^{-ivtG_{\rm SVA}^{\times}} W_v^+(z,t)] + V_1^+(z,t)$$
$$= \epsilon[e^{-ivtG_{\rm SVA}^{\times}} W_v^+(z-vt,0)] + V_1^+(z,t) \tag{6.14a}$$

and

$$V^-(z,t) = \epsilon[e^{ivtG_{\rm SVA}^{\times}} W_v^-(z+vt,0)] + V_1^-(z,t) . \tag{6.14b}$$

Equations (6.14) indicate that a short light pulse, $W_v^{\pm}(z,t)$, that propagates in a nonlinear medium, moves in space under the linear momentum operator while at the same time it undergoes a spatially dependent nonlinear distortion, expressed by the SVA momentum operator.

We note that in the quantum-mechanical formulation of nonlinear optics, because of the structure of the canonical conjugate variables, it is more convenient to describe the propagation of a polarization wave (characterized by D and B) in the nonlinear medium, rather than of an electromagnetic wave (characterized by E and B) as is often done in classical nonlinear optics. However, in most propagative experiments, light originates in free space, traverses a nonlinear medium, and is subsequently detected in free space. In such a situation, the electric and magnetic fields are continuous across the interface between two media, and are the quantities that are measured in free space. Thus, to describe a propagative experiment, the first-order polarization wave (6.8) or (6.14) should be expressed in terms of the E and B fields, so that the continuity relation can be applied between the different media to give nonlinear transmission and reflection formulas analogous to those discussed by Bloembergen and Pershan [19]. In this respect, the first-order term V_1 plays an important role in that it incorporates the coupling to the wave going in the opposite direction and thus gives rise to the nonlinear reflection. In most practical situations, however, the modification of the transmission and reflection formulas by the optical nonlinearity can be neglected, and the propagating electromagnetic wave in the nonlinear medium is approximated by the first term in Eqs. (6.8) or (6.14) which neglects all coupling to the opposite-going wave as

$$\overline{W}_v^+(z,t) = e^{itH_{1S}^{\times}} W_v^+(z,t) = e^{-ivtG_{\rm SVA}^{\times}} W_v^+(z,t) \tag{6.15a}$$

and

$$\overline{W}_v^-(z,t) = e^{itH_{1S}^{\times}} W_v^-(z,t) = e^{ivtG_{\rm SVA}^{\times}} W_v^-(z,t) . \tag{6.15b}$$

Before closing this section we shall briefly discuss the expressions for the SVA momentum operator $G_{\rm SVA}$ that are explicitly calculated in Appendix B for different nonlinear media. To make contact with the language of classical nonlinear optics, we shall express $G_{\rm SVA}$ in terms of the electric field, keeping in mind, however, that G_{SVA} is subject to the linear-medium commutator between D_0 and B_0. In each medium, G_{SVA} can be written in a form that is readily understood in terms of the well-known manifestations of the optical nonlinearity of the medium. For example, for the case of a medium with quadratic optical nonlinearity, the quadratic SVA momentum operator [Eq. (B17a)] can be written as

$$G_{\mathrm{SVA}}^{(2)}=\tfrac{1}{6}\chi^{(2)}\int E_0(B_0E_0)d\mathbf{r}+\frac{1}{6}\frac{\chi^{(2)}}{\epsilon}\int B_0\left[\frac{\epsilon E_0^2+B_0^2}{2}\right]d\mathbf{r}. \tag{6.16}$$

The integrand in the first term involves a free-field-like momentum density B_0E_0, whose "coefficient" (i.e., $\chi^{(2)}E_0/6$) can be regarded as an effective susceptibility that contributes to the refractive index. Thus the refractive index experienced by a pulse has a component that is proportional to the applied electric field, a feature referred to classically as the electro-optic (Pockels) effect. Similarly, the second term in Eq. (6.16) can be interpreted as accounting for magneto-optic phenomena. Similarly, in the presence of a cubic nonlinearity G_{SVA} [Eq. (B17b)] can be written as

$$G_{\mathrm{SVA}}^{(3)}=\frac{1}{4}\frac{\chi^{(3)}}{\epsilon}\int\left[\frac{\epsilon E_0^2+B_0^2}{2}\right]B_0E_0d\mathbf{r}\ . \tag{6.17}$$

That is, the integrand of the third-order SVA momentum operator consists of the product of two operators: the linear energy density $(\epsilon E_0^2+B_0^2)/2$ and a free-field-like momentum density B_0E_0. This form of the third-order SVA momentum operator corresponds quite well to the classical view for the treatment of the propagation of a light pulse in a Kerr medium: The nonlinear refractive index experienced by the pulse at each point (i.e., the coefficient in front of the product B_0E_0 in the SVA momentum operator) is proportional to the pulse intensity at that point (i.e., to its local energy density).

VII. AN EXAMPLE OF NONLINEAR PROPAGATION: TRAVELING-WAVE PARAMETRIC DOWN-CONVERSION

To illustrate the operatorial formalism for propagative nonlinear-optical phenomena, we shall examine the treatment of degenerate parametric down-conversion of a short pump pulse propagating in a medium that exhibits a second-order optical nonlinearity. This situation is quite often realized in experiments [20–22] on traveling-wave squeezed-light generation, whereby intense pulses from a mode-locked laser traverse a nonlinear crystal in a single-pass configuration to produce pulses of subharmonic (squeezed) light. For the case of a classical pump, this problem was treated quite early through a modal analysis by Tucker and Walls [10], and later by Lane *et al.* [23]. More recently, to overcome the shortcomings of the modal approach, Yurke *et al.* [11] and Caves and Crouch [12] treated this problem by using spatial differential equations on appropriately defined photon creation and annihilation operators.

In the calculations developed in this paper, we do not consider dispersion effects explicitly. However, dispersion is introduced implicitly by assuming that the electromagnetic field excitations can be grouped into two nonoverlapping narrow frequency bands, "pump" and "signal," coupled to each other by $\chi^{(2)}$, an assumption that simulates the spectral selectivity imposed by the phase-matching conditions of a birefringent crystal that is tuned to optimize degenerate parametric amplification. This assumption is used quite widely [11] at present. The pump pulse is assumed to be very strong compared with the parametric pulse and the distance of propagation in the nonlinear crystal is assumed to be short enough so that the pump is undepleted throughout the nonlinear process, as is the case in most experiments. The results of this section follow closely the results of the direct-space approach of Refs. [11] and [12].

As discussed in Sec. VI, an electromagnetic wave W_v^+ propagating along the positive z axis inside a nonlinear medium over a distance $z=vt$ acquires a spatial modulation envelope due to the optical nonlinearity. The full modulated wave $\overline{W}$ can be written in terms of the carrier wave W as

$$\begin{aligned}\overline{W}(z,t)&=e^{-izG_{\mathrm{SVA}}^{\times}}W(z,t)\\&=\sum_{n=0}^{\infty}\frac{(-izG_{\mathrm{SVA}}^{\times})^n}{n!}W(z,t)\ ,\end{aligned} \tag{7.1}$$

where we have omitted the subscript v and the superscript $+$ to simplify the notation. Thus, from a practical viewpoint, the description of nonlinear propagation involves essentially the calculation of multiple commutators of G_{SVA} with the (linear) displacement and magnetic-field operators of the carrier wave.

For a medium that exhibits a second-order nonlinearity, the SVA momentum operator that governs the nonlinear spatial progression is calculated in Appendix B as

$$G_{\mathrm{SVA}}^{(2)}=\frac{\chi^{(2)}}{4\epsilon}\int(B_0D_0^2+B_0^3/3)d\mathbf{r}\ . \tag{7.2}$$

Its commutator with the electric-field operator may thus be written, after a little algebra, as

$$G_{\mathrm{SVA}}^{\times}\frac{D_0}{\epsilon}=\frac{\chi^{(2)}}{\epsilon}G_0^{\times}\left[\frac{E_0^2+B_0^2/\epsilon}{2}\right]=\frac{\chi^{(2)}}{\epsilon}H_0^{\times}(B_0E_0)\ , \tag{7.3a}$$

while its commutator with the magnetic field is

$$G_{\mathrm{SVA}}^{\times}B_0=\frac{\chi^{(2)}}{\epsilon}G_0^{\times}(B_0E_0)=\chi^{(2)}H_0^{\times}\left[\frac{E_0^2+B_0^2/\epsilon}{2}\right]\ , \tag{7.3b}$$

so that in terms of the propagation of the electromagnetic wave W Eqs. (7.3) can be combined to give

$$G_{\mathrm{SVA}}^{\times}W=\frac{1}{2}\frac{\chi^{(2)}}{\epsilon}G_0^{\times}W^2=\frac{1}{2}\frac{\chi^{(2)}}{\sqrt{\epsilon}}H_0^{\times}W^2\ . \tag{7.4}$$

The advantage of expressing the commutator of the SVA momentum operator in terms of the temporal derivative of the carrier wave is that the temporal evolution of the W is identical to that of the incident field and is thus specified by the experimental conditions. In calculating the temporal derivatives, it is convenient to separate the field into its positive and negative frequency parts

$$W = 2(E^{(+)} + E^{(-)}) , \tag{7.5a}$$

where the factor of 2 is introduced so that the amplitude of $E^{(+)}$ and $E^{(-)}$ used in Eq. (7.4) coincides with that of the conventional positive- and negative-frequency electric-field operators [24]. It arises because, in our definition (4.6), W includes both the electric and magnetic fields. We note that since W is defined as a zeroth-order field operator, the positive and negative frequencies that enter in the separation of Eq. (7.5) refer to the one-particle eigenvalues of H_0. We can similarly separate the modulated wave solution $\overline{W}$ into its positive- and negative-frequency parts,

$$\overline{W} = 2(\overline{E}^{(+)} + \overline{E}^{(-)}) , \tag{7.5b}$$

where the positive and negative frequencies refer to the one-particle eigenvalues of the full Hamiltonian of the field in the nonlinear medium. In our first-order perturbative treatment, the correction to the eigenvalues is small compared with the optical frequencies. Thus, when dealing with the propagation of optical radiation, the eigenvalues (i.e., the optical frequencies) retain the same sign and Eq. (6.15) may be written as

$$\overline{E}^{(+)}(z,t) = e^{itH_{1S}^{\times}} E^{(+)}(z,t) = e^{-izG_{\mathrm{SVA}}^{\times}} E^{(+)}(z,t) , \tag{7.6}$$

with a similar equation holding for the negative-frequency part.

We now introduce the two fields involved in parametric down-conversion: the pump field, whose components oscillate at frequencies in the vicinity of the central pump frequency ω_P, and the signal field, that groups all components that oscillate at approximately ω_S such that $\omega_S = \omega_P/2$. The pump field consists of a short pulse whose duration T_P is much longer than the optical period $2\pi/\omega_P$. This implies that the temporal derivatives of the field operators can be calculated as expansions in the small parameter $\lambda_1 = 2\pi/(T_P\omega_P) \ll 1$. That is,

$$H_0^{\times} E_P^{(+)} = -\omega_P E_P^{(+)} - \lambda_1 \omega_P E_{P1}^{(+)} , \tag{7.7a}$$

where the term of order λ_1^1 is the derivative of the envelope function of the pulse, while the term of order λ_1^0 is the derivative of the carrier wave of the pulse. Similarly, for the negative-frequency part we have

$$H_0^{\times} E_P^{(-)} = \omega_P E_P^{(-)} + \lambda_1 \omega_P E_{P1}^{(-)} . \tag{7.7b}$$

Similar equations can also be derived for the other positive- and negative-frequency operators. Usually, the bandwidth of the light pulses is of the same order (or even smaller) as the gain bandwidth of the parametric process. Thus, in the calculation of the SVA momentum commutator (7.4) we only need to consider the temporal derivative of the field operators up to order $\lambda_1^{(0)}$. Equation (7.4) for the signal field thus becomes

$$G_{\mathrm{SVA}}^{\times} E_S^{(+)} = -\kappa E_P^{(+)} E_S^{(-)} , \tag{7.8a}$$

while for the negative-frequency component we have

$$G_{\mathrm{SVA}}^{\times} E_S^{(-)} = \kappa E_P^{(-)} E_S^{(+)} , \tag{7.8b}$$

where $\kappa = \chi^{(2)}\omega_S/\sqrt{\epsilon}$. Similarly, for the pump field we have

$$G_{\mathrm{SVA}}^{\times} E_P^{(+)} = -\kappa E_S^{(+)} E_S^{(+)} \tag{7.8c}$$

and

$$G_{\mathrm{SVA}}^{\times} E_P^{(-)} = \kappa E_S^{(-)} E_S^{(-)} . \tag{7.8d}$$

Since the momentum commutator is related to the spatial derivative by a Heisenberg-like equation (3.3), Eqs. (7.8) have the same form as the familiar classical first-order differential equations that describe the spatial progression of the field in a quadratic medium, within the SVA approximation [18].

In calculating the multiple commutators of G_{SVA}, we note that, for a strong undepleted pump and a weak signal field, the multiple commutators should be calculated exactly in the pump field operators, but only up to second order in the signal field operators. Thus, within the undepleted pump approximation, we have

$$\begin{aligned} G_{\mathrm{SVA}}^{\times} G_{\mathrm{SVA}}^{\times} E_S^{(+)} &= -\kappa E_P^{(+)} G_{\mathrm{SVA}}^{\times} E_S^{(-)} \\ &= -\kappa^2 (E_P^{(-)} E_P^{(+)}) E_S^{(+)} \end{aligned} \tag{7.9a}$$

and, more generally, the even-order commutators are

$$(G_{\mathrm{SVA}}^{\times})^{2n} E_S^{(+)} = (-\kappa^2)^n [(E_P^{(-)})^n (E_P^{(+)})^n] E_S^{(+)} , \tag{7.9b}$$

while for the odd-order commutators we have

$$\begin{aligned} (G_{\mathrm{SVA}}^{\times})^{2n+1} E_S^{(+)} &= (-\kappa^2)^n [(E_P^{(-)})^n (E_P^{(+)})^n] \\ &\quad \times (-\kappa E_P^{(+)}) E_S^{(-)} , \end{aligned} \tag{7.9c}$$

where we have used normal ordering for the positive- and negative-frequency components of the same central frequency (ω_P). We note that the factor inside the square brackets in the $2n$- and $(2n+1)$-order commutator is related to the equal-time normally ordered correlation function $g^{(n)}$ for the pump field, familiar from the theory of photon coincidences and photon-counting statistics [24]. This shows that the quantum statistics of the pump field conditions the propagation of the parametric signal inside the medium.

Equations (7.9) permit us to express formally the nonlinear spatial progression equation (7.1) as

$$\begin{aligned} \overline{E}_S^{(+)}(z,t) &= \sum_{n=0}^{\infty} \frac{(-izG_{\mathrm{SVA}}^{\times})^n}{n!} E_S^{(+)}(z,t) \\ &= \cosh_N[\kappa z \sqrt{I_P(z,t)}] E_S^{(+)}(z,t) \\ &\quad + i \sinh_N[\kappa z \sqrt{I_P(z,t)}] \frac{E_P^{(+)}(z,t)}{\sqrt{I_P(z,t)}} \\ &\quad \times E_S^{(-)}(z,t) , \end{aligned} \tag{7.10a}$$

where

$$I_P(z,t)=E_P^{(-)}(z,t)E_P^{(+)}(z,t) \tag{7.10b}$$

is essentially the "intensity" operator for the pump field, and $\cosh_N$ and $\sinh_N$ are normally ordered operator functions: when they or their products are expanded in a power series, the positive- and negative-frequency operators must be ordered as in Eq. (7.9).

We are now in a position to describe a traveling-wave experiment of degenerate parametric down-conversion. In such an experiment, a pump pulse, expressed by the state $|P(z,t)\rangle$, traverses a nonlinear crystal that extends from $z=0$ to L and generates a signal pulse in the course of its propagation. At the point z ($z>0$), the resulting state of the field (pump plus signal) may be written as

$$|P+S(z,t)\rangle=e^{-iG_{\mathrm{SVA}}z}|P(z,t)\rangle \ . \tag{7.11}$$

At the exit of the crystal, at $z=L$, a filter separates the pump and signal pulses and the characteristics of the signal pulse may be determined by performing different types of measurements, each expressed by the appropriate correlation function. For example, a measurement of the intensity profile of the signal pulse can be expressed by the equal-time function

$$\begin{aligned}I_S(t)&=\langle P+S(L,t)|E_S^{(-)}(L,t)E_S^{(+)}(L,t)|P+S(L,t)\rangle\\&=\langle P(0,t')|e^{iG_0L}e^{iG_{\mathrm{SVA}}L}E_S^{(-)}(L,t)E_S^{(+)}(L,t)e^{-iG_{\mathrm{SVA}}L}e^{-iG_0L}|P(0,t')\rangle \ ,\end{aligned} \tag{7.12a}$$

where

$$t'=t-L/v \ . \tag{7.12b}$$

The unitary operators that describe propagation, i.e., the carrier-wave propagation operator $\exp(-iG_0z)$ and the nonlinear modulation operator $\exp(-iG_{\mathrm{SVA}}z)$, permit us to relate the measurement of the signal field at the exit of the crystal at (L,t) to the characteristics of the incident (pump) field at the entrance of the crystal, $z=0$, and at the earlier time $t'=t-L/v$. The effect of the nonlinear envelope on the signal-field operators that describe the measurement at $z=L$ is given by an equation similar to Eq. (7.10). Thus, introducing this expression into Eq. (7.12), we can obtain an operatorial equation for the intensity profile of the signal pulse, measured at the exit of the crystal, in terms of the higher-order correlation functions (or photon-coincidence rates) for the pump wave, evaluated at the entrance of the crystal:

$$\begin{aligned}I_S(t)=&\langle P(0,t')|\sinh_N^2[\kappa L\sqrt{I_P(0,t')}]|P(0,t')\rangle\\&\times\langle 0|E_S^{(+)}E_S^{(-)}|0\rangle \ .\end{aligned} \tag{7.13}$$

This equation can be calculated for any pump wave for which the photon statistics and the temporal evolution can be modeled at the entrance of the crystal by an operatorial (nonclassical) expression. The same considerations also hold for spectral measurements that are described by the Fourier transform of the two-time correlation function

$$g_S^{(1)}(t_2,t_1)=\langle P+S|E_S^{(-)}(L,t_2)E_S^{(+)}(L,t_1)|P+S\rangle \tag{7.14}$$

or for the nth-order photon-coincidence rate for the signal pulse, which is given by an equation of the form

$$g_S^{(n)}=\langle P+S|(E_S^{(-)})^n(E_S^{(+)})^n|P+S\rangle \ . \tag{7.15}$$

A particularly simple situation occurs when the pump field corresponds to a classical (coherent) state that presents a nonzero expectation value for the electric field operator E_P, such as a laser pulse. For a laser pulse that has an amplitude profile $A_P(t')$ at $z=0$, the nth-order photon-coincidence rate is

$$g_P^{(n)}(t')=\langle P(0,t')|[E_P^{(-)}(0,t')]^n[E_P^{(+)}(0,t')]^n|P(0,t')\rangle=A_P^{2n}(t') \ . \tag{7.16}$$

Thus, according to Eq. (7.10), after the pump pulse traverses a length z in the nonlinear crystal, the signal field is characterized by the operator

$$E_S^{(+)}(z,t)=\{\cosh[\kappa zA_P(t')]E_S^{(+)}(0,t')+i\sinh[\kappa zA_P(t')]E_S^{(-)}(0,t')\}e^{-i\omega_St+ik_Sz} \ , \tag{7.17}$$

which is the operator corresponding to a short pulse of squeezed light [11,12]. Introducing this expression into Eq. (7.13), we obtain the intensity profile of the squeezed signal pulse measured at the exit of the crystal ($z=L$) as

$$I_S(L,t)=\sinh^2[\kappa LA_P(t-L/v)] \ . \tag{7.18}$$

Thus the signal pulse shape is directly related to the pump pulse profile, but this relationship changes in the

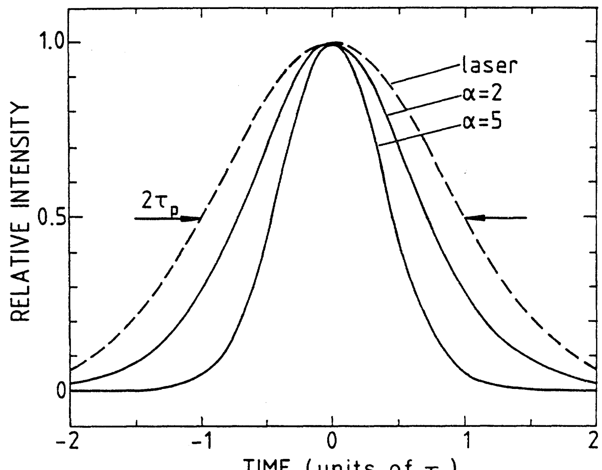

FIG. 1. Solid line, temporal profile of the intensity of a parametric signal pulse, calculated for different values of the propagation parameter $\alpha=\kappa A_p(0)z$. Dashed line, intensity profile of laser pulse. Intensity is normalized to $I(0)=1$ at the peak of each curve. Time is in units of the half width at half maximum of the laser pulse.

course of propagative parametric generation. The signal pulse is amplified as it copropagates with the pump pulse over a distance z: its peak intensity increases practically exponentially as a function of z, while at the same time its duration decreases. The shortening of the parametric signal pulse upon propagation is illustrated in Fig. 1, where we have calculated the temporal profile of the intensity of a short pulse of parametric light produced by the degenerate down-conversion of a short laser pulse of Gaussian shape for different values of the propagation parameter $\alpha=\kappa A_p(0)z$. Similarly, by calculating the Fourier transform of the two-time first-order correlation function of the signal pulse (7.14) it can be shown that the spectrum of the signal pulse broadens as it propagates in the course of its generation in the crystal.

We shall not pursue here any further the analysis of traveling-wave down-conversion of a laser pulse, as our purpose was only to illustrate that the direct-space momentum-based approach to quantum optics readily permits an analysis of temporal (or spectral) information in traveling-wave experiments, as a function of spatial progression of the electromagnetic wave.

VIII. CONCLUSIONS

The conventional theory of quantum optics deals with the spatial progression of light in an effective nonlinear medium through modal analysis, a relatively cumbersome technique for describing the nonlinear propagation of short pulses. In this paper we developed a reformulation of the effective theory of quantum optics which does not rely on a modal decomposition for the electromagnetic field, but examines directly the variation of the local electric- and magnetic-field operators in direct space. In this way, it can provide a quantum-mechanical treatment of the propagation of a short light pulse through a transparent nonlinear medium. The direct-space formulation of quantum optics uses the momentum operator of the electromagnetic field for the calculation of the spatial progression of light, in addition to the Hamiltonian that describes the temporal evolution.

A key feature of the direct-space approach to quantum optics is the derivation of the operatorial equivalent of the Maxwell equations. These equations can be combined into a single operatorial equation, analogous to the electromagnetic wave equation of classical optics. The operatorial wave equation provides a relationship between the Hamiltonian and momentum operators of the field in a (linear or nonlinear) medium, and thus permits us to describe the temporal evolution of a pulse in terms of its progression through direct space, without a modal analysis of the pulse.

Inside a nonlinear medium, the relative strength of the linear and nonlinear polarizations permits us to adopt a perturbative viewpoint and to consider the effects of the nonlinearity as a slow modulation imprinted by the medium on a fast varying carrier wave. The perturbative analysis of the operatorial wave equation up to first order yields the operatorial equivalent to the slowly-varying-amplitude equation on which is based the theory of classical nonlinear optics. This equation permits the calculation of an effective nonlinear momentum operator which can treat explicitly the spatial progression of the slow modulation envelope imprinted by the nonlinear medium on a propagating short light pulse.

Thus, the direct-space approach to quantum optics, developed in this paper, can treat nonlinear traveling-wave problems in which the quantum statistics of the propagating electromagnetic wave are important, such as the propagative parametric down-conversion of a short light pulse, or the propagation of a pulse of nonclassical light through a nonlinear medium.

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APPENDIX A: DERIVATION OF THE OPERATORIAL MAXWELL EQUATIONS

Using the equal-time commutator (2.13) between the displacement and magnetic fields

$$[D(z,t)]^{\times}B(z',t)=-i\delta'(z-z') \tag{A1}$$

we can calculate the commutator of the Hamiltonian (2.9) with the magnetic-field operator as

$$H^{\times}B(z,t)=\sum_{n=1}\beta^{(n)}\int dz' D^{n}(z',t)\{[D(z',t)]^{\times}B(z,t)\}$$
$$=-i\sum_{n=1}\beta^{(n)}\int dz' D^{n}(z',t)\delta'(z'-z)$$
$$=i\frac{\partial}{\partial z}\sum_{n=1}\beta^{(n)}D^{n}(z,t)=i\frac{\partial}{\partial z}E(z,t) \ . \tag{A2}$$

Using the Heisenberg-like equation that involves the momentum operator (3.3) Eq. (A2) may be rewritten as

$$H^{\times}B=G^{\times}E \ . \tag{A3}$$

Similarly, for the commutator of the Hamiltonian with the displacement field we have

$$H^{\times}D(z,t)=\int dz' B(z',t)\{[B(z',t)]^{\times}D(z,t)\}$$
$$=-i\int dz' B(z',t)\delta'(z'-z)=i\frac{\partial}{\partial z}B(z,t) \ , \tag{A4}$$

which, by use of Eq. (3.3), gives

$$H^{\times}D=G^{\times}B \ . \tag{A5}$$

Equations (A3) and (A5) are the operatorial equivalent of the classical Maxwell equations (2.1).

APPENDIX B: THE OPERATORS OF THE SLOWLY-VARYING-AMPLITUDE APPROXIMATION

In this appendix we calculate the operators that enter in the SVA treatment of the nonlinear wave equation, that is, the operators introduced in Eqs. (5.8), (5.19), and (6.1). We first calculate the wavelike part of the nonlinear polarization P_W, and then obtain the stationary part of the nonlinear interaction Hamiltonian H_{1S} as well as the effective SVA "momentum" operator $G_{\rm SVA}$. We express all operators in terms of the "zeroth-order" electric- and magnetic-field operators E_0 and B_0 which follow the linear-medium wave equation.

As discussed in Sec. V, we seek to partition the nth-order nonlinear polarization $P_{\rm NL}=\chi^{(n)}E_0^n$ into two parts, P_W and $(P_{\rm NL}-P_W)$, by adding or subtracting terms of the form $B_0^qE_0^r$, such that one part (P_W) is a solution of the zeroth-order (linear) wave equation (5.19)

$$(G_0^{\times}G_0^{\times}-\epsilon H_0^{\times}H_0^{\times})P_W=0 \ , \tag{B1}$$

with H_0 and G_0 being the linear-medium Hamiltonian and momentum operators, defined by Eqs. (5.2). Clearly, this partition is not unique, since any term that satisfies the zeroth-order wave equation (B1) (for example, the electric field E_0) can be added to P_W and subtracted from $P_{\rm NL}-P_W$ to give a new partition. To make the partition unique, we must add the requirement of "homogeneity," that is, that all terms added or subtracted must be of order n in the field operators. Thus all terms should be of the form $B_0^qE_0^r$ with $q+r=n$. This requirement arises from the fact that the Lagrangian density (2.3) is an effective theory that can be used exclusively at a tree level. In other words, within the effective theory any given order of $\chi^{(n)}$ should not be renormalized by the effects of the other orders.

Since the wave equation (B1) is a relationship between the temporal and spatial derivatives of P_W, we shall seek a partition of $P_{\rm NL}$ (i.e., a linear combination of $B_0^qE_0^{n-q}$) such that its commutator with the linear-medium Hamiltonian H_0 gives an exact spatial differential. To calculate this commutator, we write the operatorial Maxwell equations for a linear medium in the form

$$H_0^{\times}B_0=G_0^{\times}E_0=iE_0' \ , \tag{B2a}$$
$$H_0^{\times}E_0=\frac{1}{\epsilon}G_0^{\times}B_0=\frac{i}{\epsilon}B_0' \ , \tag{B2b}$$

where the prime denotes the spatial derivative, e.g., $E'=\partial E/\partial z$, etc. The commutator of $B_0^qE_0^r$ with the linear-medium Hamiltonian H_0 can then be calculated as

$$H_0^{\times}(B_0^qE_0^r)=i\left(qB_0^{q-1}E_0'E_0^r+\frac{r}{\epsilon}B_0^qB_0'E_0^{r-1}\right)$$
$$=i\left[\frac{q}{r+1}B_0^{q-1}(E_0^{r+1})'+\frac{r}{q+1}\frac{1}{\epsilon}(B_0^{q+1})'E_0^{r-1}\right] . \tag{B3}$$

It is now relatively easy to calculate the linear combination of $B_0^qE_0^r$ terms which upon commutation with H_0 gives an exact spatial differential. The linear combination which has E_0^n as the first term is

$$S_n=2^{-n+1}\sum_{m=0}^{n/2}\frac{n!}{(n-2m)!2m!}\epsilon^{-m}B_0^{2m}E_0^{n-2m} \tag{B4a}$$

and thus corresponds to the sum of all products $B_0^qE_0^{n-q}$ that include all possible *even* powers of B_0. The factor 2^{-n+1} is introduced for normalization and is equal to the inverse of the total number of terms in the sum that composes S_n. With this normalization S_n can be written as

$$S_n=(W_v^+/2)^n+(W_v^-/2)^n \ , \tag{B4b}$$

where W_v^+ and W_v^- are the forward and backward electromagnetic waves defined in Eqs. (4.6) and (4.8), respectively. Its commutator with H_0 can then be written as

$$H_0^{\times}S_n=\frac{1}{\epsilon}G_0^{\times}R_n=\frac{i}{\epsilon}(R_n)' \ , \tag{B5}$$

where

$$R_n=2^{-n+1}\sum_{m=0}^{n/2-1}\frac{n!}{(n-2m-1)!(2m+1)!}\epsilon^{-m}B_0^{2m+1}\times E_0^{n-2m-1}$$
$$=\sqrt{\epsilon}[(W_v^+/2)^n-(W_v^-/2)^n] \tag{B6}$$

and thus consists of the sum of all products $B_0^qE_0^{n-q}$ with all possible *odd* powers of B_0. It can be easily verified that, in addition to Eq. (B5), R_n and S_n satisfy also

$$H_0^{\times}R_n=G_0^{\times}S_n=i(S_n)' \ . \tag{B7}$$

Comparing Eqs. (B5) and (B7) with Eqs. (B2), we note that S_n and R_n transform into each other under the Maxwell equations in the same way as the electric and magnetic fields. Indeed, $S_1=E_0$ and $R_1=B_0$. Thus, combining Eqs. (B5) and (B7) it can be shown that both S_n and R_n satisfy the zeroth-order wave equation (B1). In principle, then, P_W should be expressed as a linear combination of S_n and R_n. However, since the overall polarization is a product of E fields, we choose the linear combination that itself behaves as an electric field, that is,

$$P_W^{(n)}=\chi^{(n)}(E_0^n)_W=\chi^{(n)}S_n \;. \tag{B8}$$

We note that with this choice the truncation can be written as

$$E_0^n=(W_v^+/2+W_v^-/2)^n \Longrightarrow (E_0^n)_W =(W_v^+/2)^n+(W_v^-/2)^n \tag{B9}$$

and thus corresponds to eliminating from P_{NL} all terms that couple opposite-going waves, as noted by Shen [18]. Thus the wavelike part of the second-order nonlinear polarization can be written according to Eq. (B4) as

$$(P^{(2)})_W=\chi^{(2)}\left[\tfrac{1}{2}E_0^2+\frac{1}{2\epsilon}B_0^2\right], \tag{B10a}$$

while for the third-order term we have

$$(P^{(3)})_W=\chi^{(3)}\left[\tfrac{1}{4}E_0^3+\frac{3}{4\epsilon}B_0^2E_0\right]. \tag{B10b}$$

We now look at the partition of the nonlinear interaction Hamiltonian (5.5)

$$H_1^{(n)}=-\frac{\chi^{(n)}}{n+1}\int E_0^{n+1}d\mathbf{r} \tag{B11}$$

into a stationary and a nonstationary part, as discussed in Eqs. (5.8). By adding and subtracting terms of the form $B_0^qE_0^r$, with $q+r=n+1$ for homogeneity, the stationary part of the interaction Hamiltonian H_{1S} can be written as

$$H_{1S}^{(n)}=-\frac{\chi^{(n)}}{n+1}\int S_{n+1}d\mathbf{r} \tag{B12}$$

so that for the case of a medium with quadratic nonlinearity we have

$$H_{1S}^{(2)}=-\frac{\chi^{(2)}}{3}\int\left[\tfrac{1}{4}E_0^3+\frac{3}{4\epsilon}B_0^2E_0\right]d\mathbf{r}\;, \tag{B13a}$$

while for a medium with a cubic nonlinearity we have

$$H_{1S}^{(3)}=-\frac{\chi^{(3)}}{4}\int\left[\tfrac{1}{8}E_0^4+\frac{6}{8\epsilon}B_0^2E_0^2+\frac{1}{8\epsilon^2}B_0^4\right]d\mathbf{r}\;. \tag{B13b}$$

It is relatively straightforward to show that H_{1S} commutes with both H_0 and G_0, as required by Eqs. (5.8). For H_0 we have

$$\begin{aligned} H_0^{\times}H_{1S} &= -\frac{\chi^{(n)}}{n+1}\int H_0^{\times}S_{n+1}dz \\ &= -\frac{\chi^{(n)}}{n+1}\frac{i}{\epsilon}\int (R_{n+1})'dz \\ &= -\frac{\chi^{(n)}}{n+1}\frac{i}{\epsilon}R_{n+1}\Big|_{-\infty}^{\infty}=0 \end{aligned} \tag{B14a}$$

because of the periodic boundary condition, which imposes that $R_n(-\infty)=R_n(\infty)$. It can be shown, similarly, that H_{1S} commutes with G_0. The same is true also for any operator that can be expressed as an integral over all space:

$$\begin{aligned} G_0^{\times}H_{1S} &= -\frac{\chi^{(n)}}{n+1}\int G_0^{\times}S_{n+1}dz \\ &= -\frac{\chi^{(n)}}{n+1}i\int (S_{n+1})'dz=-\frac{\chi^{(n)}}{n+1}iS_{n+1}\Big|_{-\infty}^{\infty}=0 \end{aligned} \tag{B14b}$$

because of the periodic boundary condition.

We may similarly calculate the effective SVA "momentum" operator, defined by Eq. (6.1) as

$$G_{\mathrm{SVA}}^{\times}(\epsilon E_0)=\tfrac{1}{2}G_0^{\times}P_W\;. \tag{B15}$$

Using the partition of P_{NL} that corresponds to Eqs. (B8) and (B9), for the case of a medium with an nth-order nonlinearity, $G_{\mathrm{SVA}}^{(n)}$ can be written as

$$G_{\mathrm{SVA}}^{(n)}=\frac{\chi^{(n)}}{n+1}\int R_{n+1}d\mathbf{r} \tag{B16}$$

as can be readily verified by introducing Eq. (B16) into Eq. (B15) and using the explicit expressions for S_n and R_n, (B4) and (B6), respectively. Clearly, $G_{\mathrm{SVA}}^{(n)}$ given by Eq. (B16) commutes with both H_0 and G_0, as can be shown in a way analogous to Eqs. (B14).

For the particular case of a medium with a quadratic nonlinearity, Eq. (B16) gives

$$G_{\mathrm{SVA}}^{(2)}=\frac{\chi^{(2)}}{4}\int\left[B_0E_0^2+\frac{1}{3\epsilon}B_0^3\right]d\mathbf{r}\;, \tag{B17a}$$

while for a medium with a cubic nonlinearity we have

$$G_{\mathrm{SVA}}^{(3)}=\frac{\chi^{(3)}}{8}\int\left[B_0E_0^3+\frac{1}{\epsilon}B_0^3E_0\right]d\mathbf{r}\;. \tag{B17b}$$

[1] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Photons and Atoms: Introduction to Quantum Electrodynamics* (Wiley, New York, 1989).

[2] J. M. Jauch and K. M. Watson, Phys. Rev. **74**, 950 (1948).

[3] Y. R. Shen, Phys. Rev. **155**, 921 (1967).

[4] R. J. Glauber and M. Lewenstein, in *Squeezed and Nonclassical Light*, edited by P. Tombesi and E. R. Pike, Vol. 190 of *NATO Advanced Study Institute, Series B: Physics*

(Plenum, New York, 1989), p. 203; R. J. Glauber and M. Lewenstein, Phys. Rev. A **43**, 467 (1991).

[5] M. Hillery and L. D. Mlodinow, Phys. Rev. A **30**, 1860 (1984).

[6] P. D. Drummond and S. J. Carter, J. Opt. Soc. Am. B **4**, 1565 (1987).

[7] B. Yurke, Phys. Rev. A **29**, 408 (1984); **32**, 300 (1985).

[8] M. J. Collett and C. W. Gardiner, Phys. Rev. A **30**, 1386 (1984); C. W. Gardiner and M. J. Collett, *ibid.* **31**, 3761 (1985).

[9] H. J. Carmichael, J. Opt. Soc. Am. B **4**, 1565 (1987).

[10] J. Tucker and D. F. Walls, Phys. Rev. **178**, 2036 (1969).

[11] B. Yurke, P. Grangier, R. E. Slusher, and M. J. Potasek, Phys. Rev. A **35**, 3586 (1987).

[12] C. M. Caves and D. D. Crouch, J. Opt. Soc. Am. B **4**, 1535 (1987).

[13] I. Abram, Phys. Rev. A **35**, 4661 (1987).

[14] I. Abram, in *Photons and Quantum Fluctuations,* edited by E. R. Pike and H. Walther, *Malvern Physics Series* (Hilger, Bristol, 1988), p. 173.

[15] E. A. Power and S. Zienau, Philos. Trans. R. Soc. London, Ser. A **251**, 427 (1959); R. G. Woolley, Proc. R. Soc. London Ser. A **321**, 557 (1971); M. Babiker and R. Loudon, *ibid.* **385**, 439 (1983).

[16] C. Itzykson and J. B. Zuber, *Quantum Field Theory* (McGraw-Hill, New York, 1980).

[17] R. Kubo, J. Phys. Soc. Jpn. **17**, 1100 (1962).

[18] Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley-Interscience, New York, 1984).

[19] N. Bloembergen and P. Pershan, Phys. Rev. **128**, 606 (1962).

[20] I. Abram, R. K. Raj, J. L. Oudar, and G. Dolique, Phys. Rev. Lett. **57**, 2516 (1986).

[21] R. E. Slusher, P. Grangier, A. LaPorta, B. Yurke, and M. J. Potasek, Phys. Rev. Lett. **59**, 2566 (1987).

[22] P. Kumar, O. Aytur, and J. Huang, Phys. Rev. Lett. **64**, 1015 (1990).

[23] A. Lane, P. Tombesi, H. J. Carmichael, and D. F. Walls, Opt. Commun. **48**, 155 (1983).

[24] R. J. Glauber, Phys. Rev. **130**, 2529 (1963).