Emission of photon pairs at discontinuities of nonlinearity in spontaneous parametric down-conversion

Jan Peřina Jr., Antonín Lukš, Ondřej Haderka
Joint Laboratory of Optics of Palacký University and Institute of Physics of Academy of Sciences of the Czech Republic, 17. listopadu 50A, 772 07 Olomouc, Czech Republic

In order to fulfil the continuity requirements for electric- and magnetic-field amplitudes at discontinuities of $\chi^{(2)}$ nonlinearity additional photon pairs have to be emitted in the area of discontinuity. Generalized two-photon spectral amplitudes can be used to describe properties of photon pairs generated in this process that we call surface spontaneous parametric down-conversion. The spectral structure of such photon pairs is similar to that derived for photon pairs generated in the volume. Surface and volume contributions to spontaneous down-conversion can be comparable as an example of nonlinear layered structures shows.

I. INTRODUCTION

The generation of second-harmonic field at a boundary between two homogeneous media with different values of $\chi^{(2)}$ nonlinearity has been addressed for the first time more than 30 years ago $\cite{1,2}$. This weak effect has been discovered when second-harmonic generation with considerable phase mismatch has been investigated. The surface second-harmonic field occurs here naturally and assures the fulfilment of continuity requirements for the tangential components of electric- and magnetic-field vector amplitudes that stem from Maxwell’s equations. In more detail, a fundamental field creates a nonlinear polarization at second-harmonic frequency at the nonlinear side of the boundary. This polarization generates two surface second-harmonic fields, one in forward direction, one in backward direction. As a consequence two different second-harmonic fields propagate inside a nonlinear crystal. They differ in their wave vectors. The first (and usual) field originates from the volume nonlinear polarization and its local wave vector is twice the wave vector of the fundamental field. On the other hand the wave vector given by index of refraction at second-harmonic frequency characterizes the second-harmonic field arising at the boundary and propagating freely through the crystal. Experimental evidence of these effects can be found, e.g., in $\cite{2}$. Surface second-harmonic generation pumped by ultrashort pulses has been analyzed in $\cite{3}$. Deep understanding of this effect can be reached when studying this process in a nonlinear medium with negative index of refraction $\cite{4}$. In Ref. $\cite{4}$, completely numeric approach based on the solution of nonlinear Maxwell’s equations has been adopted contrary to the original analytical and approximate approach in $\cite{1}$ demonstrating the richness of physical effects included implicitly in Maxwell’s equations. We note that also absorption in highly phase-mismatched volume second-harmonic generation has been observed $\cite{5}$.

The above described effects are valid for nonlinear parametric (three-mode) interactions in which a large number of material states far from resonance participate. On the other hand resonant second-harmonic generation mediated by resonant surface states has been widely studied for many materials (see, e.g., in $\cite{6,7}$) and has become a useful tool for surface diagnostics at present. We note that also entangled photon pairs generated in parametric down-conversion can be converted resonantly into plasmons at material surfaces even with the preservation of polarization entanglement $\cite{8}$.

The question arises whether nonresonant surface effects can occur also in the quantum process of spontaneous parametric down-conversion (SPDC) $\cite{9}$. In the case of second-harmonic generation, the presence of macroscopic classical nonlinear polarization at frequency $2\omega$ is crucial. On the other hand, there is no macroscopic classical nonlinear polarization at the signal- or idler-field frequencies. However, quantum nonlinear polarization occurs in SPDC at these frequencies and is responsible for photon-pair generation at a boundary.

SPDC is described by an appropriate momentum operator that is constructed in the framework of energy-flux quantization $\cite{10,11,12}$. We note that the inclusion of all fields occurring during the propagation, i.e. forward- as well as backward-propagating fields, is necessary to keep consistency of the approach.

The article is divided as follows. A model of surface SPDC in case of a homogeneous nonlinear crystal is developed in Sec. II. Determination of physical quantities characterizing photon pairs is described in Sec. III using generalized two-photon amplitudes. Sec. IV gives a generalization to nonlinear layered structures. Conclusions are drawn in Sec. V.

II. MOMENTUM OPERATOR AND FIELDS’ CONTINUITY AT THE BOUNDARIES OF A NONLINEAR CRYSTAL

In this section, we first pay attention to the volume nonlinear interaction, then study the problem at the input and later at the output boundaries and finally add the obtained expressions describing the photon-pair generation. We note that a simplified model has been presented in $\cite{13}$. 
A. Volume interaction

The following interaction momentum operator \( \hat{G}_{\text{int}} \) is appropriate for the process of SPDC \[9, 11\]:

\[
\hat{G}_{\text{int}}(z) = 4\epsilon_0 A \int dt \sum_{\alpha,\beta,\gamma=F,B} d_{\gamma,\alpha}\times \left[ E_{p\gamma}^{(+)}(z,t) \hat{E}_{\alpha}^{(-)}(z,t) \hat{E}_{\gamma}^{(-)}(z,t) + \text{h.c.} \right],
\]

(1)

where \( E_{p\gamma}^{(+)} \) are positive-frequency parts of the (linearly polarized) pump-field electric-field amplitudes whereas \( \hat{E}_{\alpha}^{(-)} \) (\( \hat{E}_{\gamma}^{(-)} \)) stand for negative-frequency parts of the signal- (idler-) field electric-field amplitude operators. Subscript \( F \) (\( B \)) refers to a field propagating forward (backward), i.e. along the +\( z \) (−\( z \)) axis. Symbol \( \epsilon_0 \) means permittivity of vacuum, \( d_{\gamma,\alpha\beta} \) are effective nonlinear coefficients, \( A \) is transverse area of the fields, and h.c. replaces the hermitian-conjugated terms. We have assumed a scalar model for the interacting fields for simplicity. However, a generalization to the vectorial model is straightforward because of the applied first-order perturbation approximation. Using a spectral decomposition of the interacting fields,

\[
E_{m\alpha}(z,t) = \frac{1}{\sqrt{2\pi}} \int d\omega m E_{m\alpha}^{(+)}(z,\omega m) \exp(-i\omega m t),
\]

(2)

the momentum operator \( \hat{G}_{\text{int}} \) in Eq. (1) can be recast into the form

\[
\hat{G}_{\text{int}}(z) = \frac{4\epsilon_0 A}{\sqrt{2\pi}} \int d\omega m \int d\omega s \int d\omega i \delta(\omega_p - \omega_s - \omega_i) \times \left[ E_{p\gamma}^{(+)}(z,\omega_p) \hat{E}_{s\alpha}^{(-)}(z,\omega_s) \hat{E}_{i\beta}^{(-)}(z,\omega_i) + \text{h.c.} \right],
\]

(3)

where the \( \delta \)-function expresses conservation of energy for monochromatic waves. The signal and idler spectral electric-field amplitude operators \( \hat{E}_{s\alpha}^{(-)} \) can be expressed in terms of creation operators \( \hat{a}_{m\alpha}^{\dag} \) introduced such that \( \hat{a}_{m\alpha}^{\dag} \hat{a}_{m\alpha} \) gives the photon-number density in mode \( m \alpha \) at a given frequency:

\[
\hat{E}_{s\alpha}^{(-)}(z,\omega m) = -i \sqrt{\frac{\hbar \omega m}{2\epsilon_0 c A n_m (\omega m)}} \hat{a}_{m\alpha}^{\dag}(z,\omega m),
\]

(4)

\[
m = s, i, \quad \alpha = F, B.
\]

Symbol \( n_m \) stands for an index of refraction of field \( m \). Spatial evolution of optical fields is determined by the solution of Heisenberg equations \[15, 10\] for field operators denoted as \( \hat{X} \):

\[
\frac{d\hat{X}(z)}{dz} = -\frac{i}{\hbar} \left[ \hat{G}(z), \hat{X}(z) \right];
\]

(5)

\[
\hat{G}(z) = \hat{G}_0(z) + \hat{G}_{\text{int}}(z),
\]

\[
\hat{G}_0(z) = \sum_{m=s,i} \sum_{\alpha=F,B} \hbar \int d\omega m k_{m\alpha}(\omega m) \times \hat{a}_{m\alpha}^{\dag}(z,\omega m) \hat{a}_{m\alpha}(z,\omega m);
\]

(6)

the interaction momentum operator \( \hat{G}_{\text{int}} \) is given in Eq. (1). The momentum operator \( \hat{G}_0 \) introduced in Eq. (6) describes free-field evolution. Symbol \( k_{m\alpha} \) stands for a wave vector of mode \( m \alpha \) at frequency \( \omega_m \); \( k_{mp} = k_m, k_{mp} = -k_m, k_m > 0 \). Symbol \( \hbar \) denotes the reduced Planck constant.

In order to determine the electric-field amplitude operators at the output of the nonlinear crystal, the Heisenberg equations for operators \( \hat{a}_{m\alpha}(z,\omega_m) \) \( m = s, i, \alpha = F, B \) have to be solved:

\[
\frac{d\hat{a}_{s\alpha}(z,\omega_m)}{dz} = i k_{s\alpha}(\omega_s) \hat{a}_{s\alpha}(z,\omega_m) + \sum_{\beta,\gamma=F,B} \int d\omega i g_{\gamma,\alpha\beta}(\omega_s,\omega_i) E_{p\gamma}^{(+)}(0,\omega_s + \omega_i) \times \exp[i k_{p\gamma}(\omega_s + \omega_i) z] \hat{a}_{i\beta}^{\dag}(z,\omega_i), \quad \alpha = F, B.
\]

(7)

Equations (7) have been derived assuming equal-space commutation relations \[15\]. We note that fields propagating along the −\( z \) axis have negative wave-vectors in the definition of the free-field momentum operator \( \hat{G}_0 \) in Eq. (6). A more detailed and rigorous formulation of the dynamics of counter-propagating fields justifying this approach can be found in \[13\]. The coupling constants \( g_{\gamma,\alpha\beta} \) occurring in Eq. (7) are given along the expression:

\[
g_{\gamma,\alpha\beta}(\omega_s,\omega_i) = \frac{2i d_{\gamma,\alpha\beta}}{\epsilon c \sqrt{2\pi n_s(\omega_s) n_i(\omega_i)}},
\]

(8)

Equations for operators \( \hat{a}_{i\beta} \) and \( \hat{a}_{i\beta} \) can be derived from Eq. (7) by the formal substitution \( s \rightarrow i \).

Solution of Eq. (7) valid up to the first power of \( g \) can be obtained in the form:

\[
\hat{a}_{s\alpha}(z,\omega_s) = \exp[i k_{s\alpha}(\omega_s) z] \hat{a}_{s\alpha}(0,\omega_s) + \sum_{\beta,\gamma=F,B} \int d\omega i B_{\gamma,\alpha\beta}(z,\omega_s,\omega_i) \hat{a}_{i\beta}^{\dag}(0,\omega_i),
\]

(9)

where

\[
B_{\gamma,\alpha\beta}(z,\omega_s,\omega_i) = g_{\gamma,\alpha\beta}(\omega_s,\omega_i) E_{p\gamma}^{(+)}(0,\omega_s + \omega_i) \times \exp[i \Delta k_{\gamma,\alpha\beta}(\omega_s,\omega_i) z/2] \times z \text{sinc}[\Delta k_{\gamma,\alpha\beta}(\omega_s,\omega_i) z/2];
\]

(10)

\[
\Delta k_{\gamma,\alpha\beta}(\omega_s,\omega_i) = k_{p\gamma}(\omega_s + \omega_i) - k_{s\alpha}(\omega_s) - k_{i\beta}(\omega_i); \quad \alpha, \beta, \gamma = F, B.
\]

(11)

\text{sinc}(x) = \sin(x)/x. \text{ We note that the restriction to the first power of } g \text{ in the formula in Eq. (9) has justified the use of the approximate formula } \hat{a}_{m\alpha}(z,\omega_m) =
\[ \exp[i k_{m_n}(\omega_m) z] \hat{a}_{m_n}(0, \omega_m) \] below the integral over the frequency \( \omega_1 \). The solution in Eq. (4) describes SPDC originating in the volume of nonlinear crystal.

The solution obtained for annihilation operators \( \hat{a}_{m_n}(z, \omega_m) \) as written in Eq. (4) provides the following expressions for the positive-frequency parts of electric- \( (\hat{E}_{s_n}^+) \) and magnetic-field \( (\hat{H}_{s_n}^+) \) amplitude operators:

\[
\hat{E}_{s_n}^+(z, \omega_s) = i \sqrt{\frac{\hbar \omega_s}{2 \epsilon_0 c A_n(\omega_s)}} \exp[i k_{s_n}(\omega_s) z] \sum_{\beta, \gamma = F, B} A_{\beta, \gamma}(z, \omega_s, \omega_1) \hat{a}_{\beta, \gamma}^\dagger(0, \omega_1),
\]

(12)

\[
\hat{H}_{s_n}^+(z, \omega_s) = \hat{H}_{s_n}^{+\text{Fr}}(z, \omega_s) + \hat{H}_{s_n}^{+\text{Fr}}(z, \omega_s),
\]

(13)

\[
\hat{H}_{s_n}^{+\text{Fr}}(z, \omega_s) = \frac{\hbar c}{2 \mu_0 z c A_n(\omega_s)} H_{s_n}(\omega_s)
\]

\[
\times \sum_{\beta, \gamma = F, B} \int d\omega_i g_{\beta, \gamma}(\omega_s, \omega_i) E_{p_\beta}^+(\omega_s + \omega_i) \exp[i k_{p_\beta}(\omega_s + \omega_i) z] \hat{a}_{\beta, \gamma}^\dagger(0, \omega_i),
\]

(14)

Equations (13–15) for the magnetic-field amplitude operators \( \hat{H}_{s_n}^+ (\alpha = F, B) \) have been derived assuming polarization of electric-field amplitudes \( \hat{E}_{s_n}^+ \) along the +x axis and, consequently, polarization of magnetic-field amplitudes \( \hat{H}_{s_n}^+ \) along the +y axis. The Maxwell equations then provide the following formula \( \hat{H}_{s_n}^+ = -i/\omega_0 \mu_0 \partial \hat{E}_{s_n}^+ / \partial z \), where \( \mu_0 \) stands for permeability of vacuum. The magnetic-field amplitude operators \( \hat{H}_{s_n}^+(z, \omega_s) \) have been decomposed in Eq. (13) into two parts; the amplitude operators \( \hat{H}_{s_n}^{+\text{Fr}}(z, \omega_s) \) are linearly proportional to the electric-field amplitude operators \( \hat{E}_{s_n}^+(z, \omega_s) \) whereas the amplitude operators \( \hat{H}_{s_n}^{+\text{nFr}}(z, \omega_s) \) are of purely nonlinear origin. The amplitude operators \( \hat{H}_{s_n}^{+\text{nFr}} \) are not taken into account in the usual derivation of Fresnel’s relations that assumes linear media. Correct inclusion of these amplitude operators into the continuity considerations at a boundary results in additional contributions to the nonlinear process.

**B. Input boundary**

Let us take deeper attention to the problem of continuity of electric- and magnetic-field amplitudes at the boundaries of the nonlinear medium. First we pay attention to the input boundary \( (z = 0) \) and consider the signal field. Four electric- and magnetic-field amplitudes are involved in the continuity requirement: amplitudes \( E_{s_F}(0) \) and \( H_{s_F}(0) \) of the forward-propagating field impinging on the boundary from the outside of nonlinear medium, amplitudes \( E_{s_B}(0) \) and \( H_{s_B}(0) \) leaving the boundary outside the nonlinear crystal, amplitudes \( E_{s_B}(0) \) and \( H_{s_B}(0) \) leaving the boundary and propagating inside the nonlinear crystal [see Fig. 1]. Because the magnetic-field amplitudes \( H_{s_F}(0) \) and \( H_{s_B}(0) \) defined in the nonlinear crystal have also nonlinear contributions \( H_{s_F}^\text{nFr}(0) \) and \( H_{s_B}^\text{nFr}(0) \) described in Eq. (15) additional (surface) amplitude corrections \( \delta E_{s_F}(0) \) and \( \delta E_{s_B}(0) \) together with \( \delta H_{s_F}(0) \) and \( \delta H_{s_B}(0) \) are needed to fulfil the continuity requirement. The surface amplitude corrections naturally occur in the fields that leave the boundary which is a consequence of spatio-temporal considerations that are suppressed to certain extent in our one-dimensional model.

The requirement of continuity of projections of electric- and magnetic-field amplitudes to the plane of the input boundary leads to the following equations:

\[
E_{s_F}(0) + E_{s_B}(0) + \delta E_{s_F}(0) = E_{s_F}(0) + \delta E_{s_F}(0),
\]

(16)

\[
H_{s_F}(0) + H_{s_B}(0) + \delta H_{s_F}(0) = H_{s_F}^\text{Fr}(0) + H_{s_F}^\text{nFr}(0),
\]

(17)

Derivation of the usual Fresnel’s relations (in linear media) [22] is based on the fulfilment of the following equations:

\[
E_{s_F}(0) + E_{s_B}(0) = E_{s_F}(0) + E_{s_B}(0),
\]

(18)

\[
H_{s_F}(0) + H_{s_B}(0) = H_{s_F}^\text{Fr}(0) + H_{s_F}^\text{nFr}(0).
\]

(19)

Comparison of Eqs. (16) and (18) together with Eqs. (17) and (19) results in two algebraic equations for the surface amplitude corrections of the fields leaving the boundary:

\[
\delta E_{s_F}(0) = \delta E_{s_F}(0),
\]

(20)

\[
\delta H_{s_F}(0) = H_{s_F}^\text{nFr}(0) + \delta H_{s_F}(0) + H_{s_F}^\text{nFr}(0).
\]

(21)

Alternatively and more conveniently, the amplitude corrections \( \delta E_{s_B} \) and \( \delta H_{s_B} \) of the field outside the nonlinear crystal can be formally included into the equations giving Fresnel’s relations. This can be done if we introduce fictitious amplitude corrections \( \delta E_{s_B} \) and \( \delta H_{s_B} \) of the field impinging on the boundary from its nonlinear side. Such corrections give, after transformation at the boundary using Fresnel’s relations, the required amplitude corrections \( \delta E_{s_B} \) and \( \delta H_{s_B} \) [see Fig. 1]. Then we have:

\[
E_{s_F}(0) + \left[ E_{s_B}(0) + \delta E_{s_B}(0) \right] = E_{s_F}(0)
\]
Solution of Eqs. (28) and (29) finally gives the expressions for annihilation-operator corrections $\delta a_{s_F}$ and $\delta a_{s_B}$ at the input boundary:

$$
\delta a_{s_F}(0, \omega_i) = \sum_{\alpha, \beta, \gamma = F, B} \int d\omega_l \, g_{\gamma, \alpha \beta}(\omega_s, \omega_l) \times E_{p\gamma}^{(+)}(\omega_s + \omega_l) \hat{a}^\dagger_{i \beta}(0, \omega_l) \tag{30}
$$

$$
\delta a_{s_B}(0, \omega_i) = \delta a_{s_F}(0, \omega_i). \tag{31}
$$

C. Output boundary

Fields at the output boundary of the nonlinear crystal can be analyzed similarly as at the input boundary. Here, the continuity of projections of electric- and magnetic-field amplitudes at $z = L$ ($L$ denotes the crystal length) to the plane of the boundary gives two equations:

$$
E_{s_F}(L) + E_{s_B}(L) + \delta E_{s_B}(L) = E_{s_F}^{(1)}(L) + \delta E_{s_F}^{(1)}(L), \tag{32}
$$

$$
H_{s_F}^{Fr}(L) + H_{s_B}^{Fr}(L) + H_{s_F}^{Fr}(L) + H_{s_B}^{Fr}(L) + \delta H_{s_B}(L) = H_{s_F}^{(1)}(L) + \delta H_{s_F}^{(1)}(L) + H_{s_B}^{(1)}(L). \tag{33}
$$

Amplitudes $E_{s_F}^{(1)}(L)$ and $H_{s_F}^{(1)}(L)$ describe the field outside the nonlinear crystal whereas amplitudes $E_{s_B}^{(1)}(L)$ and $H_{s_B}^{(1)}(L)$ refer to the field impinging on the output boundary from its linear side (see Fig. 1). The amplitude corrections $\delta E_{s_F}^{(1)}(L)$ and $\delta H_{s_F}^{(1)}(L)$ can be formally included into the equations that express Fresnel’s relations provided that fictitious amplitude corrections $\delta E_{s_F}(L)$ and $\delta H_{s_F}(L)$ are introduced. Motivation for this step is the same as in the case of input boundary: we want to have corrections only inside the nonlinear crystal. We then have:

$$
[E_{s_F}(L) + \delta E_{s_F}(L)] + E_{s_B}(L) = E_{s_F}^{(1)}(L) + \delta E_{s_F}^{(1)}(L), \tag{34}
$$

$$
[H_{s_F}^{Fr}(L) + \delta H_{s_F}(L)] + H_{s_B}^{Fr}(L) = H_{s_F}^{(1)}(L) + \delta H_{s_F}^{(1)}(L) + H_{s_B}^{(1)}(L). \tag{35}
$$

Comparison of Eqs. (34) and (35) with Eqs. (32) and (33) results in two algebraic equations for the surface amplitude corrections inside the nonlinear medium:

$$
- \delta E_{s_F}(L) + \delta E_{s_B}(L) = 0, \tag{36}
$$

$$
H_{s_F}^{Fr}(L) - H_{s_F}(L) + H_{s_B}^{Fr}(L) + \delta H_{s_B}(L) = 0. \tag{37}
$$

Two algebraic equations for the annihilation-operator corrections $\delta \hat{a}_{s_F}(L, \omega_s)$ and $\delta \hat{a}_{s_B}(L, \omega_s)$ can be derived from Eqs. (30) and (31) using the expressions in Eqs.
\[ -\delta a_{sF}(L,\omega_s) + \delta a_{sB}(L,\omega_s) = 0, \]  
\[ -ik_s(\omega_s)\delta a_{sF}(L,\omega_s) + ik_s(\omega_s)\delta a_{sB}(L,\omega_s) + \sum_{\alpha,\beta,F,B} \int d\omega_i g_{\gamma,\alpha\beta}(\omega_s,\omega_i) \times E^{(+)}_p(\omega_s + \omega_i) \exp[ik_p(\omega_s + \omega_i)L] \times \exp[-ik_{i\beta}(\omega_i)L]a^\dagger_{i\beta}(0,\omega_i) = 0. \]  
\[
\delta a_{sF}(L,\omega_s) = -i \frac{2k_s(\omega_s)}{\delta k_L(\omega_s)} \sum_{\alpha,\beta,F,B} \int d\omega_i g_{\gamma,\alpha\beta}(\omega_s,\omega_i) \times E^{(+)}_p(\omega_s + \omega_i) \exp[ik_p(\omega_s + \omega_i)L] \times \exp[-ik_{i\beta}(\omega_i)L]a^\dagger_{i\beta}(0,\omega_i),
\]
\[
\delta a_{sB}(L,\omega_s) = \delta a_{sF}(L,\omega_s).
\]

D. The whole nonlinear crystal

The overall solution for operators \( \hat{a}_{s\alpha}(L,\omega_s) \) valid up to the first power of \( g \) has three nonlinear contributions: the first comes from the input boundary, the second from the volume and the third from the output boundary. The overall solution can be written as follows:

\[
\hat{a}_{s\alpha}(L,\omega_s) = \hat{a}^\text{free}_{s\alpha}(L,\omega_s) + \sum_{\beta,\gamma,F,B} \int d\omega_i \mathcal{F}_{\gamma,\alpha\beta}^s(L,\omega_s,\omega_i) \hat{a}^\text{free}_{i\beta}(L,\omega_i), \quad \alpha = F,B.
\]

Operators \( \hat{a}^\text{free}_{m\alpha}(L,\omega_m) \) (\( \alpha = s,i \), \( \alpha = F,B \)) occurring in Eqs. \( 12 \) and \( 13 \) below and expressed at the crystal end correspond to free-field evolution (i.e., without photon-pair generation) inside the crystal. They are determined by the formula \( \hat{a}^\text{free}_{m\alpha}(L,\omega_m) = \exp[ik_{m\alpha}(\omega_m)L]a^\text{free}_{m\alpha}(0,\omega_m) \). Functions \( \mathcal{F}_{\gamma,\alpha\beta}^s \) are defined in Eqs. \( 14 \) and \( 15 \) below.

Now we pay attention to the idler fields and use symmetry between the signal and idler fields. The idler-field electric- (magnetic-) field amplitudes are assumed to be polarized along the \(+y\) (\(-x\)) axis. The requirement of continuity of electric- and magnetic-field amplitudes at the input and output boundaries leads to the solution for idler-field operators \( \hat{a}_{i\beta}(L,\omega_i) \) in the form:

\[
\hat{a}_{i\beta}(L,\omega_i) = \hat{a}_{i\beta}^\text{free}(L,\omega_i) + \sum_{\alpha,\gamma,F,B} \int d\omega_s \mathcal{F}_{\gamma,\alpha\beta}^i(L,\omega_s,\omega_i) \hat{a}^\text{free}_{s\alpha}(L,\omega_s), \quad \beta = F,B.
\]

The functions \( \mathcal{F}^s \) and \( \mathcal{F}^i \) introduced in Eqs. \( 12 \) and \( 13 \) can be decomposed into volume (\( \mathcal{F}^\text{vol} \)) and surface (\( \mathcal{F}^\text{surf} \)) contributions:

\[
\mathcal{F}_{\gamma,\alpha\beta}^m = \mathcal{F}_{\gamma,\alpha\beta}^\text{vol} + \mathcal{F}_{\gamma,\alpha\beta}^\text{surf},
\]

\[
\mathcal{F}_{\gamma,\alpha\beta}^\text{surf}(L,\omega_s,\omega_i) = \frac{\Delta k_{\gamma,\alpha\beta}(\omega_s,\omega_i)}{k_m(\omega_m)} \times \exp[ik_{p\beta}(\omega_s + \omega_i)L] \times \exp[ik_{i\alpha}(\omega_i)L] \times \exp[ik_{p\alpha}(\omega_s + \omega_i)L] \times \exp[ik_{i\beta}(\omega_i)L],
\]

\[ m = s,i; \quad \alpha,\beta,\gamma = F,B. \]

In deriving Eq. \( 16 \) we have assumed \( g_{\gamma,F,B} = g_{\gamma,FB} \) and \( g_{\gamma,F} = g_{\gamma,FB} \). The functions \( \mathcal{F}^\text{surf} \) describing surface contributions disappear in the limit \( L \to 0 \), i.e. the surface contribution from the output boundary completely compensates that from the input boundary. Comparison of the expressions in Eqs. \( 15 \) and \( 16 \) reveals a simple relation between the volume and surface contributions:

\[
\mathcal{F}_{\gamma,\alpha\beta}^\text{surf}(L,\omega_s,\omega_i) = \mathcal{F}_{\gamma,\alpha\beta}^\text{vol}(L,\omega_s,\omega_i) \Rightarrow \mathcal{F}_{\gamma,\alpha\beta}^\text{surf}(L,\omega_s,\omega_i) = \mathcal{F}_{\gamma,\alpha\beta}^\text{vol}(L,\omega_s,\omega_i).
\]

The structure of surface contribution is formed by mutual interference of fields generated at the input and output boundaries. At a boundary, only the energy conservation restricts properties of an emitted photon pair. Such photon pair thus has a rich internal spectral structure because phase-matching conditions do not apply. It is then the mutual interference of fields coming from the input and output boundaries that gives conditions similar to those of phase matching naturally found in the volume interaction.

In standard bulk sources of photon pairs that are typically several mm long, the interaction among forward-propagating pump, signal, and idler fields is important. In this case, the commonly-used formalism for the description of SPDC [using a two-photon spectral amplitude \( \Phi(\omega_s,\omega_i) \)] can be applied (see, e.g., \( 17 \) \( 18 \) \( 19 \)). The surface contributions can be involved in this formalism if the following formal substitution is done:

\[
\Phi(\omega_s,\omega_i) \leftrightarrow \sqrt{1 + \mathcal{F}^\text{surf}_F(\omega_s,\omega_i)} \times \sqrt{1 + \mathcal{F}^\text{surf}_F(\omega_s,\omega_i)} \Phi^\text{vol}(\omega_s,\omega_i),
\]

where the two-photon amplitude \( \Phi^\text{vol}(\omega_s,\omega_i) \) characterizes the usual volume contribution. Under the usual condition of phase-matched nonlinear interaction \( [\Delta k_{FF}(\omega_s,\omega_i)] = 0 \) the surface contributions occur only at spectral tails and are negligible.
Surface contributions have typically broader spectra compared to the volume contributions. If these spectra are not filtered in an experimental setup they lead to sharper features of two-photon temporal amplitudes $\tilde{F}^{s,\text{surf}}(\tau_s, \tau_i)$ and $\tilde{F}^{i,\text{surf}}(\tau_s, \tau_i)$ [for their definition, see Eq. (50) below]. As documented in Fig. 2b, for a BBO crystal, the surface two-photon temporal amplitudes $\tilde{F}_m^{n,\text{surf}}$ $(m = s, i)$ attain large values in the vicinity of the input and output boundaries. On the other hand the two-photon temporal amplitude $\tilde{F}^{\text{vol}}$ of the volume contribution has roughly the same values along the whole nonlinear crystal. If the volume SPDC is strongly phase mismatched its two-photon temporal amplitude $\tilde{F}^{\text{vol}}$ resembles that of the surface SPDC (see Fig. 2b). This means that only values of the two-photon temporal amplitude $\tilde{F}^{\text{vol}}$ characterizing photon pairs born in the vicinity of crystal edges are higher. Destructive interference inside the nonlinear crystal prevails in this case and suppresses photon-pair emission. Because the signal- and idler-field spectra are very broad for phase-mismatched volume SPDC is similar to that appropriate for surface SPDC. Alternatively, sum-frequency interactions for operators $\hat{a}_{s, F}$, $\hat{a}_{i, B}$, $\hat{a}_{i, F}$, and $\hat{a}_{i, B}$ requires a generalization of the usually used formulas for the determination of physical quantities characterizing photon pairs. For simplicity we pay attention to photon pairs with both photons propagating forward and define operators $\hat{a}_{m}(\omega_m) \equiv t_m(\omega_m)\hat{a}_{m,F}(L, \omega_m)$ and $\hat{E}_{m}^{(+)}(\tau_m) \equiv t_m(\omega_m)\hat{E}_{m,F}^{(+)}(L, \omega_m)$, where the coefficients $t_m$ describe the amplitude transmissivities at the output boundary $(m = s, i)$. We have $t_m = 2n_m/(n_m + n_m^{(1)})$ according to Fresnel’s formulas, $n_m(n_m^{(1)})$ means an index of refraction of (beyond) the nonlinear medium $(m = s, i)$.

**FIG. 2:** Cross-section of the absolute value of two-photon temporal amplitude $F^s(\tau_s) \equiv |\tilde{F}_{F,F,F}^s(\tau_s, 0)|$ for the surface ($b = s$, surf, solid line with *) and volume ($b = v$, vol, solid line) contributions assuming cw pumping. The values of $|\tilde{F}_{F,F,F}^s|$ are 5000 (10) times magnified with respect to those of $|\tilde{F}_{F,F,F}^v|$ in case a (b). The curves are appropriate for a BBO crystal 5 mm long and collinear type-II interaction at the pump wavelength of 400 nm and signal and idler wavelengths 800 nm (frequency filters 30 nm wide (FWHM) are used). The crystal optical axis declines by 43.25 deg (perfect phase matching) (a) and 80 deg (b) with respect to the axis of fields’ propagation.

**III. QUANTITIES CHARACTERIZING THE EMITTED PHOTON PAIRS**

The inclusion of surface contributions to the expressions for operators $\hat{a}_{i,F}$, $\hat{a}_{i,B}$, $\hat{a}_{i,F}$, and $\hat{a}_{i,B}$ requires a generalization of the usually used formulas for the determination of physical quantities characterizing photon pairs. For simplicity we pay attention to photon pairs with both photons propagating forward and define operators $\hat{a}_{m}(\omega_m) \equiv t_m(\omega_m)\hat{a}_{m,F}(L, \omega_m)$ and $\hat{E}_{m}^{(+)}(\tau_m) \equiv t_m(\omega_m)\hat{E}_{m,F}^{(+)}(L, \omega_m)$, where the coefficients $t_m$ describe the amplitude transmissivities at the output boundary $(m = s, i)$. We have $t_m = 2n_m/(n_m + n_m^{(1)})$ according to Fresnel’s formulas, $n_m(n_m^{(1)})$ means an index of refraction of (beyond) the nonlinear medium $(m = s, i)$. 
The joint signal-idler photon-number density \( n(\omega_s, \omega_i) \) giving the number of emitted photon pairs with a signal photon in the unit interval around frequency \( \omega_s \) and its idler twin in the unit interval around frequency \( \omega_i \) at the medium output plane is defined as follows:

\[
n(\omega_s, \omega_i) = \left\langle \left[ \hat{a}_s^\dagger(\omega_s)\hat{a}_s(\omega_s)\hat{a}_i^\dagger(\omega_i)\hat{a}_i(\omega_i) + \text{h.c.} \right] \right\rangle / 2,
\]

\( (49) \)

Symbol \( \langle \rangle \) means averaging over the initial signal-and-idler-field vacuum states. Substitution of Eqs. (42) and (43) into the definition of the joint signal-idler-photon-number density \( n \) in Eq. (49) results in the formula:

\[
n(\omega_s, \omega_i) = \text{Re}\{\tilde{F}^s(\omega_s, \omega_i)\tilde{F}^i(\omega_s, \omega_i)\}.
\]

\( (50) \)

The functions \( \tilde{F}^s \) and \( \tilde{F}^i \) in Eq. (50) include transmission through the output boundary:

\[
\tilde{F}^m(\omega_s, \omega_i) = t_s(\omega_s)t_i(\omega_i)\tilde{F}^s_F,F_F^\text{p}(\omega_s, \omega_i), \quad m = s, i.
\]

\( (51) \)

In Eq. (50), symbol \( \text{Re} \) denotes the real part of an argument. We note that expressions containing factor \( \delta^2(\omega_p) \) occur in formulas like that written in Eq. (50) for cw pumping. This factor has to be replaced using the formula \( \delta^2(\omega_p) = 2T/(2\pi)\delta(\omega_p) \) in this case where \( 2T \) means the detection-interval length \( T \).

Intensity spectrum \( S_s(\omega_s) \) of, e.g., the signal field can be easily derived from the joint photon-number density \( n \):

\[
S_s(\omega_s) = \hbar \omega_s \int_0^\infty d\omega_i \, n(\omega_s, \omega_i)
= \hbar \omega_s \int_0^\infty d\omega_i \, \text{Re}\{\tilde{F}^s(\omega_s, \omega_i)\tilde{F}^i(\omega_s, \omega_i)\}.
\]

\( (52) \)

Photon flux of, e.g., the signal photons \( N_s(\tau_s) \) beyond the nonlinear medium can be derived along the following formula considering only photons emitted in photon pairs:

\[
N_s(\tau_s) = \epsilon_0 \mathcal{C}_s^{(1)}(\omega_s^0)A \int d\omega_i \left\langle \left[ \hat{E}_s^-(\tau_s)\hat{E}_s^+(\tau_s)\hat{a}_s^\dagger(\omega_i)\hat{a}_s(\omega_i) + \text{h.c.} \right] \right\rangle. \quad (53)
\]

Using the solutions in Eqs. (42) and (43) we arrive at the expression:

\[
N_s(\tau_s) = \frac{\hbar}{2\pi} \int d\omega_s \sqrt{\omega_s} \int d\omega_i \sqrt{\omega_i} \int d\omega_i \text{Re}\{\exp[i(\omega_s - \omega_i)\tau_s]\tilde{F}^s(\omega_s, \omega_i)\tilde{F}^s(\omega_s, \omega_i)\}. \quad (54)
\]

Assuming a narrow idler-field spectrum we can rearrange the formula in Eq. (54) as follows:

\[
N_s(\tau_s) = \hbar \omega_s \int d\tau_s \text{Re}\{\tilde{F}^s(\tau_s, \tau_s)\tilde{F}^s(\tau_s, \tau_s)\}. \quad (55)
\]

The functions \( \tilde{F}^s \) and \( \tilde{F}^i \) in time domain in Eq. (55) are derived from their spectral counterparts given in Eqs. (51), (53), and (54) along the formula:

\[
\tilde{F}^m(\tau_s, \tau_i) = \frac{1}{2\pi} \int d\omega_s \int d\omega_i \sqrt{\omega_s \omega_i} \tilde{F}^m(\omega_s, \omega_i) \times \exp(-i\omega_s\tau_s) \exp(-i\omega_i\tau_i), \quad m = s, i. \quad (56)
\]

The number \( N \) of coincidence counts caused by a simultaneous detection of both photons from one pair in a Hong-Ou-Mandel interferometer is given as:

\[
N(\tau_s) = \epsilon_0^2 \mathcal{C}_s^{(1)}(\omega_s^0)\mathcal{C}_i^{(1)}(\omega_i^0)A^2 \times \int dt_1 \int dt_2 \left\{ \left| |r|^2 + |t|^2 \right| \left\langle \left[ \hat{E}_s^-(t_1) \hat{E}_s^+(t_1) \hat{E}_i^-(t_2) \hat{E}_i^+(t_2) + \text{c.c.} \right] \right\rangle + \left\langle \left( r^* t \right)^2 \hat{E}_s^-(t_1) \hat{E}_s^+(t_2) \hat{E}_i^-(t_2 - \tau - \tau_1) \hat{E}_i^+(t_2 - \tau - \tau_1) + \text{c.c.} \right\rangle \right\}; \quad (57)
\]

\( r \) (t) stands for amplitude reflectivity (transmissivity) of a beam-splitter in the interferometer and \( \tau \) denotes a relative time delay introduced between the signal and idler photons. Using the solutions in Eqs. (42) and (43) the normalized coincidence-count number \( R_n \) can be derived in the form:

\[
R_n(\tau_s) = 1 - \rho(\tau_s), \quad (58)
\]

where

\[
\rho(\tau_s) = \frac{1}{4R_0} \int d\omega_s \int d\omega_i \text{Re}\{\left( r^* t \right)^2 \times \tilde{F}^s(\omega_s, \omega_i) \tilde{F}^i(\omega_s, \omega_i) \exp[i(\omega_s - \omega_i)\tau_s] \} \quad (59)
\]

and

\[
R_0 = \frac{1}{8} \left( |r|^4 + |t|^4 \right) \int d\omega_i \int d\omega_s \text{Re}\left\{ \tilde{F}^s(\omega_s, \omega_i) \tilde{F}^i(\omega_s, \omega_i) \right\}. \quad (60)
\]
IV. NONLINEAR LAYERED STRUCTURES

We now consider structures composed of both linear and nonlinear layers with thicknesses typically in hundreds of nm. Fulfilment of phase-matching conditions is not important in these short layers, because $\Delta k L \ll \pi$ ($L$ means a typical layer length). Surface SPDC becomes important in these structures because volume contributions to SPDC from individual layers are weak. A generalization of the developed theory to layered structures is straightforward and is based on the fact that we detect only one photon pair. This pair is generated in one of the nonlinear layers and propagates to the output of the structure. The theory giving volume contributions to SPDC has been developed in \[23, 24, 25\] using a perturbation solution to Schrödinger equation. Here, we restrict ourselves to a scalar model and collinear interaction to emphasize the important steps in the derivation. A generalization including general directions of fields’ propagation and their polarization properties is straightforward following the way presented in \[23\].

First we generalize the momentum operator $\hat{G}_{\text{int}}$ written in Eq. (3) to include the nonlinear interaction in $N$ layers:

$$\hat{G}_{\text{int}}(z) = \frac{4\varepsilon_0 A}{\sqrt{2\pi}} \int d\omega_p \int d\omega_s \int d\omega_i \delta(\omega_p - \omega_s - \omega_i) \sum_{l=1}^{N} \sum_{\alpha,\beta=\gamma=F,B} d_{\gamma,\alpha\beta}^{(l)}$$

$$\times \left[ E_{p_i}^{(+)(l)}(z,\omega_p)\hat{E}_{s_{\alpha}}^{(-)(l)}(z,\omega_s)\hat{E}_{i\beta}^{(-)(l)}(z,\omega_i) + \text{h.c.} \right],$$

where the amplitude operators $\hat{E}_{m_{\alpha}}^{(-)(l)}$ are defined in $l$th layer:

$$\hat{E}_{m_{\alpha}}^{(-)(l)}(z,\omega_m) = -i \sqrt{\frac{\hbar \omega_m}{2\varepsilon_0 c A n_m^{(l)}(\omega_m)}} \text{rect}_{z_{l-1},z_l}(z)$$

$$\times g_{m\alpha}^{(l)}(z,\omega_m), \quad m = s, i, \quad \alpha = F, B. \quad (61)$$

The pump-field spectral amplitude $E_{p_i}^{(+)(l)}$ ($\gamma = F, B$) is defined inside the $l$th layer similarly as the amplitudes in Eq. (23). The nonlinear coefficients $d_{\gamma,\alpha\beta}^{(l)}$ as well as indices of refraction $n_m^{(l)}(\omega_m)$ characterize the $l$th layer that extends from $z_{l-1}$ to $z_l$. Function $\text{rect}_{a,b}(z)$ equals 1 for $a < z < b$ and is zero otherwise.

Photon-pair generation in the $l$th layer can be studied using the approach and results presented in Sec. II. These formulas give us appropriate operators $\hat{a}_{m\alpha}(z_l,\omega_m)$ ($m = s, i; \quad \alpha = F, B$) at the end of the $l$th layer. Fresnel’s relations at the boundaries have to be used to ‘transfer’ a generated photon pair outside the boundaries of the layered structure. A photon pair can be emitted in any of $N$ layers and the corresponding quantum trajectories have to be superposed. So we can write:

$$\hat{a}^{\text{out}}_{m_{\alpha}}(\omega_m) = \sum_{l=1}^{N} \sum_{\alpha^*=F,B} T_{\alpha\alpha^*}^{m_{\alpha}}(\omega_m)\hat{a}^{(l)}_{m_{\alpha}}(z_l,\omega_m),$$

$$m = s, i, \quad \alpha , \alpha^* = F, B. \quad (63)$$

The coefficients $T_{\alpha\alpha^*}^{m_{\alpha}}$ in Eq. (63) can be derived using the propagation matrix method and Fresnel’s relations at boundaries \[23, 24\]. Properties of photon pairs as described by quantities introduced in Sec. III and measured by a simultaneous detection of both photons comprising a photon pair can be derived from the fourth-order correlation function $\langle \hat{a}_s^{\text{out}(l)}(\omega_s')\hat{a}_s^{\text{out}(l)}(\omega_s)\hat{a}_i^{\text{out}(l)}(\omega_i')\hat{a}_i^{\text{out}(l)}(\omega_i) \rangle$. Substituting Eq. (63) into the definition of the correlation function and using the solution for one nonlinear layer as presented in Eqs. (12) and (13) we arrive at:

$$\langle \hat{a}_s^{\text{out}(l)}(\omega_s')\hat{a}_s^{\text{out}(l)}(\omega_s)\hat{a}_i^{\text{out}(l)}(\omega_i')\hat{a}_i^{\text{out}(l)}(\omega_i) \rangle = \mathcal{F}_{\alpha\alpha'^*}^{\text{out}(l)}(\omega_s',\omega_s)\mathcal{F}_{\beta\beta'^*}^{\text{out}(l)}(\omega_i',\omega_i)$$

$$\times \mathcal{F}_{\alpha'^*\beta'^*}^{\text{out}(l)}(\omega_s,\omega_i), \quad o = s, i; \quad \alpha, \beta, \alpha'^*, \beta'^* = F, B. \quad (64)$$

Expressions in Eqs. (14) and (17) can be rearranged into the form:

$$\mathcal{F}_{\alpha\beta}^{\text{out}(l)}(\omega_s,\omega_i) = \sum_{\gamma=F,B} \sum_{\alpha'^*, \beta'^*} g_{\gamma,\alpha\beta}^{(l)}(\omega_s,\omega_i) \left[ 1 + \mathcal{V}_{\gamma,\alpha\beta}^{\text{out}(l)}(\omega_s,\omega_i) \right]$$

$$\times E_{p_i}^{(+)(l)}(\omega_s + \omega_i) \exp[ik_{\alpha\beta}^{(l)}(\omega_s + \omega_i)L_l/2]$$

$$\times \exp[-i\Delta k_{\gamma,\alpha\beta}^{(l)}(\omega_s,\omega_i)L_l/2], \quad o = s, i; \quad \alpha, \beta = F, B, \quad (65)$$

where

$$\mathcal{V}_{\gamma,\alpha\beta}^{\text{out}(l)}(\omega_s,\omega_i) = \frac{\Delta k_{\gamma,\alpha\beta}^{(l)}(\omega_s,\omega_i)}{k_{\alpha\beta}^{(l)}(\omega_o)},$$

$$o = s, i; \quad \alpha, \beta, \gamma = F, B \quad (67)$$

and $g_{\gamma,\alpha\beta}^{(l)}(\omega_s,\omega_i) = 2id_{\gamma,\alpha\beta}^{(l)}\sqrt{\omega_s\omega_i}/[2\pi c n_{\gamma}^{(l)}(\omega_s)n_{\gamma}^{(l)}(\omega_i)]$. Superscript $(l)$ denotes quantities appropriate for the $l$th layer and $L_l$ means the length of this layer ($L_l = z_l - z_{l-1}$). Symbol $E_{p_i}^{(+)(l)}$ occurring in Eq. (60)
refers to the spectral amplitude of field $p_\gamma^{} (\gamma = F, B)$ at the end of the $l$th layer (at $z = z_1$) and can be determined using the propagation matrix formalism. In deriving Eq. (66), we have assumed that $g_{r,FB}^{(l)} = g_{r,B\beta}^{(l)}$ and $g_{r,\alpha F}^{(l)} = g_{r,\alpha B}^{(l)}$.

The signal-field intensity spectrum $S_s$, its photon flux $N_s$, and coincidence-count interference rate $R_n$ in the Hong-Ou-Mandel interferometer can then be determined using the formulas in Eqs. (52), (51), and (50) using the spectral two-photon amplitudes $f_s^{x,\text{out}}$ and $f_s^{f,\text{out}}$ defined in Eq. (65).

The generalization including fields’ propagation under nonzero angles of incidence is straightforward following the procedure presented above in Sec. II. The key point here is that the necessary amplitude corrections assuring the fulfilment of continuity requirements for the electric and magnetic fields at the boundaries are defined only at one side of the boundaries. To be more specific, we use nonzero amplitude-operator corrections $\delta a_s p (0, \omega_s)$ and $\delta a_s p (0, \omega_i)$ $[\delta a_s p (L, \omega_s)$ and $\delta a_s p (L, \omega_i)]$ for the left-hand [right-hand] side of the nonlinear medium (layer) to describe surface SPDC. When nonzero angles of incidence are considered, we can argue as follows. The angle of incidence of a given field equals (up to the sign) the angle of reflection and so their cosines giving multiplicative factors for projections of electric- and magnetic-field amplitudes (lying in the plane of incidence) to the plane of boundary are the same. This means that the equations written in Eqs. (28), (29), (38), and (39) remain valid for any angle of incidence. Equations (66) and (67) can then be used in this case provided that we use the $z$ components of wave vectors $k_p, k_s$, and $k_i$ instead of their full lengths.

Surface SPDC can significantly contribute to the number of the generated photon pairs in layered structures. As an example, we consider a structure composed of 24 layers of GaN of the thickness of 117 nm that sandwich 24 linear layers of AlN of the thickness of 180 nm. This structure is a source of photon pairs has been studied in detail in [23, 24] for $s$-polarized pump (normally-incident), signal, and idler beams and the pump wavelength of 664.5 nm. Considering only volume contributions efficient photon-pair generation occurs at degenerate frequencies of the signal and idler fields for the signal-field emission angle of 14 deg (see Fig. 5 in [23]). The surface contributions lead to additional non-negligible photon-pair generation, as documented in Fig. 4 where the ratio $S_s^{FF,\text{vol+surf}} / S_s^{FF,\text{vol}}$ of signal-field spectral intensities with $(S_s^{FF,\text{vol+surf}})$ and without $(S_s^{FF,\text{vol}})$ surface contributions is plotted as a function of signal-field emission angle $\theta_s$. This ratio is minimum under the conditions giving the best constructive interference (i.e., when $\omega_s = \omega_i^{0}/2$ and $\theta_s = 14$ deg) and equals approximately 2. This means that the surface contributions roughly double the number of emitted photon pairs. It should be emphasized that constructive interference between the volume and surface contributions plays the key role here. Comparison of the graph in Fig. 1 with that plotted in Fig. 5 in [23] indicates that the worse the constructive interference inside the layered structure the greater the relative contribution of surface terms. However, the overall number of generated photon pairs is quite low in this region.

The increase of photon-pair generation rate is caused mainly by processes that do not (even roughly) obey phase matching conditions. Weights of the surface contributions for different processes can be judged according to the value of parameter $\gamma_{m,\alpha \beta}^{(l)}$ ($m = s, i; \alpha, \beta, \gamma = F, B$) defined in Eq. (67). The following values are met in GaN for the studied structure around the point where the best constructive interference has been found: $\gamma_{m,FB}^{FF} \approx 2\gamma_{m,F_B} \approx 0.05, \gamma_{m,F_F} \approx 2\gamma_{m,B_F} \approx 0.05$ and $\gamma_{m,B_F} \approx 2, \gamma_{m,B_B} = \gamma_{m,B_F}$. Because the dominant role in surface SPDC is played by the highly phase-mismatched processes, lengths of nonlinear layers have to be less or comparable to the coherence length of the nonlinear interaction to observe these contributions. The ratio $N_{FF,\text{surf}} / N_{FF,\text{vol}}$ of the numbers of emitted photon pairs in a certain spectral region coming from the surface $(N_{FF,\text{surf}})$ and volume $(N_{FF,\text{vol}})$ contributions for one layer of GaN as plotted in Fig. 6 indicates that the inclusion of surface contributions is important for the lengths below 1 $\mu$m.

In general, the inclusion of surface contributions leads to the broadening of spectral two-photon amplitudes $f_s^{x,\text{out}} (\omega_s, \omega_i)$ and $f_s^{f,\text{out}} (\omega_s, \omega_i)$. Thus widths of the signal- and idler-field intensity spectra also increase. The Schmidt decomposition of two-photon spectral amplitudes reveals that entropy of its coefficients increases provided that the surface terms are included. This means that entanglement between the signal and idler fields increases due to surface SPDC. On the other hand and considering pulsed pumping, photon fluxes $N$ and coincidence-count interference rates $R_n$ in a Hong-Ou-
FIG. 5: Ratio $N_{FF,\text{surf}}/N_{FF,\text{vol}}$ of the numbers of emitted photon pairs originating in surface ($N_{FF,\text{surf}}$) and volume ($N_{FF,\text{vol}}$) down-conversion from one layer of GaN of length $L$ pumped at $\lambda_p = 664.5$ nm at normal incidence. Pairs with frequencies in the interval $\Delta\Omega$ used in the graph in Fig. 4 and propagating along the $+z$ axis are collected; $N = \int_{\Delta\Omega} d\omega_s \int_{\Delta\Omega} d\omega_i n(\omega_s, \omega_i)$ and $n$ is given in Eq. (49). Logarithmic scale on the $y$ axis is used.

FIG. 6: Signal-field photon flux $N_{FF}^s$ including volume (solid line with *), surface (solid line with $\triangle$), and volume + surface (solid line with $\circ$) contributions. The structure is pumped by a Gaussian pulse with the duration of 250 fs (for details, see [23]).

Mandel interferometer as temporal characteristics become narrower as documented in Figs. 3 and 7. The overall photon flux $N$, e.g., the pulsed signal field occurs earlier at the output of the nonlinear structure compared to the case of only the volume interaction because the structure is ‘less-resonant’ (‘less-transparent’) for the surface contributions than for the volume ones. Phase modulation of the two-photon spectral amplitude characterizing the surface contributions provides the coincidence-count pattern $R_n(\tau_l)$ in the form of a global dip with visibility equal to 1 and two small side-dips (see Fig. 7).

Surface effects in SPDC occur at discontinuities of $\chi^{(2)}$ nonlinearity. Such discontinuities are found also in periodically-poled nonlinear crystals like LiNbO$_3$. In these crystals, an enhancement of photon-pair generation rates is expected provided that the period of poling is sufficiently short so that a sufficiently large number of discontinuities is present inside the nonlinear crystal. The surface effects are also expected in wave-guiding geometries where the conditions of total reflection of fields at the boundaries are met. This might be important mainly in photonic fibers. The studied surface effects are by no means restricted to 1D geometries, even stronger surface contributions might arise from 2D or 3D nonlinear structures. An effective enhancement of the nonlinear interaction caused by the surface effects should also be observed in stimulated $\chi^{(2)}$ processes like second-harmonic generation.

V. CONCLUSIONS

Surface spontaneous parametric down-conversion has been predicted combining the solution of quantum Heisenberg equations and the continuity requirements at boundaries. Formulas for the determination of the number of generated photon pairs, spectra, and photon-fluxes of the down-converted fields as well as coincidence-count rates in a Hong-Ou-Mandel interferometer have been derived using generalized signal- and idler-field two-photon spectral amplitudes. It has been shown that surface contributions from the input and output boundaries of a nonlinear crystal give structures similar to those characterizing the volume contributions. Nevertheless they are weak. The surface contributions are important whenever strongly phase-mismatched nonlinear interactions give considerable contributions, e.g., in nonlinear layered structures. An example of a GaN/AlN structure composed of several tens of layers has shown that the surface and volume contributions can be comparable in their amplitudes. This shows that the role of surface effects in other nonlinear structures like periodically-poled materials, nonlinear wave-guiding structures or structures with stimulated processes should be addressed.
Acknowledgments

Support by projects IAA100100713 of GA AV ČR, MSM6198959213, COST 09026, 1M06002, and AVOZ 10100522 of the Czech Ministry of Education is acknowledged. J.P. and O.H. thank M. Scalora and E. Fazio for discussion about surface second-harmonic generation.

[1] N. Bloembergen and P.S. Pershan, Phys. Rev. 128, 606 (1962).
[2] N. Bloembergen, H.J. Simon, and C.H. Lee, Phys. Rev. 181, 1261 (1969).
[3] M. Mlejnek, E.M. Wright, J.V. Moloney, and N. Bloembergen, Phys. Rev. Lett. 83, 2934 (1999).
[4] V. Roppo, M. Centini, C. Sibilia, M. Bertolotti, D. de Ceglia, M. Scalora, N. Akozbek, M.J. Bloemer, J.W. Haus, O.G. Kosareva, and V.P. Kandidov, Phys. Rev. A 76, 033829 (2007).
[5] M. Centini, V. Roppo, E. Fazio, F. Pettazzi, C. Sibilia, J.W. Haus, J.V. Foreman, N. Akozbek, M.J. Bloemer, and M. Scalora, Phys. Rev. Lett. 101, 113905 (2008).
[6] B.S. Mendoza and W.L. Mochán, Phys. Rev. B 53, 4999 (1996).
[7] B.S. Mendoza, A. Gaggiotti, and R. Del Sole, Phys. Rev. Lett. 81, 3781 (1998).
[8] E. Altewischer, M.F. van Exter, and J.P. Woerdman, Nature 418, 304 (2002).
[9] L. Mandel and E. Wolf, Optical Coherence and Quantum Optics (Cambridge University Press, Cambridge, 1995), chap. 22.4.
[10] R.W. Boyd, Nonlinear Optics (Academic Press, Amsterdam, 2003), 2nd edition.
[11] B. Huttner, S. Serulnik, Y. Ben-Aryeh, Phys. Rev. A 42, 5594 (1990).
[12] A. Lukš and V. Peřínová, Progress in Optics Vol. 43, Ed. E. Wolf (Elsevier, Amsterdam, 2002), p. 295.
[13] W. Vogel, D.G. Welsch, and S. Walentowicz, Quantum Optics (Wiley-VCH, Weinheim, 2001).
[14] J. Peřina Jr., A. Lukš, O. Haderka, and M. Scalora, Phys. Rev. Lett. 103, 063902 (2009).
[15] J. Peřina Jr. and J. Peřina, in Progress in Optics Vol. 41, Ed. E. Wolf (Elsevier, Amsterdam, 2000), p. 361.
[16] J. Peřina, Quantum Statistics of Linear and Nonlinear Optical Phenomena (Kluwer, Dordrecht, 1991).
[17] T. E. Keller and M. H. Rubin, Phys. Rev. A 56, 1534 (1997).
[18] W. P. Grice, R. Erdmann, I. A. Walmsley, D. Branning, Phys. Rev. A 57, R2289 (1998).
[19] J. Peřina Jr., A. V. Sergienko, B. M. Jost, B. E. A. Saleh, M. C. Teich, Phys. Rev. A 59, 2359 (1999).
[20] S. E. Harris, Phys. Rev. Lett. 98, 063602 (2007).
[21] M. B. Nasr, S. Carrasco, B. E. A. Saleh, A. V. Sergienko, M. C. Teich, J. P. Torres, L. Torner, D. S. Hum, M. M. Fejer, Phys. Rev. Lett. 100, 183601 (2008).
[22] M. Born and E. Wolf, Principles of Optics (Pergamon Press, Oxford, 1980), 6th edition.
[23] J. Peřina Jr., M. Centini, C. Sibilia, M. Bertolotti, and M. Scalora, Phys. Rev. A 73, 033823 (2006).
[24] M. Centini, J. Peřina Jr., L. Sciscione, C. Sibilia, M. Scalora, M.J. Bloemer, and M. Bertolotti, Phys. Rev. A 72, 033806 (2005).
[25] J. Peřina Jr., M. Centini, C. Sibilia, M. Bertolotti, and M. Scalora, Phys. Rev. A 75, 013805 (2007).
[26] P. Yeh, Optical Waves in Layered Media (Wiley, New York, 1988).