Surface spontaneous parametric down-conversion

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Surface spontaneous parametric down-conversion is predicted as a consequence of continuity requirements for electric- and magnetic-field amplitudes at a discontinuity of $\chi^{(2)}$ nonlinearity. A generalization of the usual two-photon spectral amplitude is suggested to describe this effect. Examples of nonlinear layered structures and periodically-poled nonlinear crystals show that surface contributions to spontaneous down-conversion can be important.

When studying the process of second-harmonic generation under considerable phase mismatch more than thirty years ago, the generation of second-harmonic field from a boundary between two homogeneous media that differ by values of $\chi^{(2)}$ nonlinearity has been discovered [1, 2]. The surface second-harmonic field arises here as a consequence of continuity requirements for projections of electric- and magnetic-field vector amplitudes into the plane of the boundary. Physically, a pumping field at frequency $\omega$ creates a step profile of nonlinear polarization at frequency $2\omega$ and with wave vector $2k(\omega)$ that becomes the source of the usual volume second-harmonic field. The wave vector of the surface second-harmonic field is $k(2\omega)$ in agreement with dispersion properties of the nonlinear material. This effect is even found in nonlinear media with negative index of refraction as the numerical solution of nonlinear Maxwell equations revealed in [3]. The studied parametric effect should be distinguished from resonant surface second-harmonic generation.

Spontaneous parametric down-conversion (SPDC) [4] belongs together with second-harmonic generation to $\chi^{(2)}$ processes. This poses the question about surface effects in SPDC. In volume SPDC, photon pairs are generated from the quantum state, due to quantum fluctuations (or quantum noise) inherent in this state. In this case, a nonlinear material responds to the presence of optical fields through quantum nonlinear polarization that acts as a source of new fields. In a close vicinity of the boundary, the interacting fields as well as the nonlinear polarization are modified in order to comply with natural fields’ continuity requirements at the boundary. This results in the generation of additional photon pairs from the area of the boundary (several wavelengths thick) that constitute surface SPDC.

Our study of surface SPDC is organized as follows. Nonlinear Heisenberg equations are derived first to treat SPDC inside the nonlinear medium. Nonlinear corrections to electric- and magnetic-field amplitudes occur naturally at boundaries and give additional, i.e. surface, contributions to SPDC. Subsequently, the derivation of quantities characterizing the emitted photon pairs is addressed. Finally, two important examples are discussed.

Adopting the quantization of energy flux [5, 6] we describe the process of SPDC involving the signal, idler, and pump fields by the Heisenberg equations with an appropriate interaction momentum operator $\hat{G}_{\text{int}}$ [3]:

$$\hat{G}_{\text{int}}(z) = \frac{4\epsilon_0 d_{\text{eff}} A}{\sqrt{2\pi}} \sum_{\alpha,\beta,\gamma=F,B} \int d\omega_s \int d\omega_i \left[ E^{(-)}_{\alpha s}(z,\omega_s + \omega_i) E^{(+)}_{\beta s}(z,\omega_s) E^{(+)}_{\gamma i}(z,\omega_i) + \text{h.c.} \right].$$ (1)

The positive-frequency part of an electric-field amplitude $E^{(+)}_{\alpha s}(z,\omega)$ can be expressed using annihilation operator $\hat{a}_{m\alpha}$ as follows $(m = p, s, i; \alpha = F, B)$:

$$\hat{E}^{(+)}_{m\alpha}(z,\omega_m) = i \sqrt{\frac{\hbar \omega_m}{2\epsilon_0 c A n_m(\omega_m)}} \hat{a}_{m\alpha}(z,\omega_m);$$ (2)

$$\hat{E}^{(-)}_{m\alpha} = (\hat{E}^{(+)}_{m\alpha})^\dagger.$$ Subscript $F$ (B) indicates a field propagating forward (backward), i.e. along $+z$ ($-z$) axis. Symbol $\epsilon_0$ means permittivity of vacuum, $d_{\text{eff}}$ is effective nonlinear coefficient, $A$ transverse area of the fields, $c$ speed of light in vacuum, and h.c. replaces the hermitian-conjugated terms. Symbol $k_m$, is a wave vector, $\omega_m$ frequency, and $n_m$ index of refraction of field $m\alpha$.

The Heisenberg equations, e.g., for the signal-field operators $\hat{a}_{s\alpha}(z,\omega_s)$ can then be derived assuming equal-space commutation relations [3]:

$$\frac{d\hat{a}_{s\alpha}(z,\omega_s)}{dz} = ik_{s\alpha}(\omega_s) \hat{a}_{s\alpha}(z,\omega_s)$$

$$+ \sum_{\beta,\gamma=F,B} \int d\omega_i g(\omega_s,\omega_i) E^{(+)}_{p\beta}(0,\omega_s + \omega_i) \times \exp(ik_{p\beta}(\omega_s + \omega_i)z) \hat{a}_{i\gamma}^\dagger(z,\omega_i), \quad \alpha = F, B;$$ (3)

Coupling constant $g$, $g(\omega_s,\omega_i) = 2\epsilon_0 d_{\text{eff}} \sqrt{\omega_s \omega_i}/(c\sqrt{2\pi}) \sqrt{n_s(\omega_s)n_i(\omega_i)}$, is linearly proportional to nonlinear coefficient $d_{\text{eff}}$.

The solution of Eq. (3) for annihilation operators $\hat{a}_{s\alpha}(z,\omega_s)$ up to the first power of $g$ gives us the formula for operator $\hat{E}^{(+)}_{s\alpha}$ defined in Eq. (2):

$$\hat{E}^{(+)}_{s\alpha}(z,\omega_s) = i \sqrt{\frac{\hbar \omega_s}{2\epsilon_0 c A n_s(\omega_s)}} \exp(ik_{s\alpha}(\omega_s)z)$$

$$\times \sum_{\beta,\gamma=F,B} \int d\omega_i g(\omega_s,\omega_i) E^{(+)}_{p\beta}(0,\omega_s + \omega_i) \times \exp(ik_{p\beta}(\omega_s + \omega_i)z) \hat{a}_{i\gamma}^\dagger(z,\omega_i), \quad \alpha = F, B;$$ (3)

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\[
\times \left[ \hat{a}_{s, \alpha}(0, \omega_s) + \sum_{\beta, \gamma = F, B} \int d\omega \, g(\omega_s, \omega_i) \right.
\times E_{\beta}^{(+)}(0, \omega_s + \omega_i) \exp[\Delta k_{\beta, \alpha}^s(\omega_s, \omega_i)z/2]
\times z \sin[\Delta k_{\beta, \alpha}^s(\omega_s, \omega_i)z/2]a_{\beta}^\dagger(0, \omega_i) \Bigg] ; \quad \alpha = F, B; (4)
\]

\[
sinc(x) = \sin(x)/x \quad \text{and} \quad \Delta k_{\beta, \alpha}^s(\omega_s, \omega_i) = k_{0, \beta}^s(\omega_s + \omega_i) - k_{0, \alpha}^s(\omega_i).
\]

The positive-frequency magnetic-field amplitude operators \(\hat{H}_{s, \alpha}^{(+)}\) can be derived using the formula
\[
\hat{H}_{s, \alpha}^{(+)}(z, \omega_s) = -i/(\omega_s \mu_0) \partial \hat{E}_{s, \alpha}^{(+)}(z, \omega_s)/\partial z \quad (\mu_0 \text{ denotes permeability of vacuum}) \text{ provided that the electric-field [magnetic-field] amplitude } E_{s, \alpha}[H_{s, \alpha}] \text{ is polarized along +x [+y] axis. The obtained operator } \hat{H}_{s, \alpha}^{(+)} \text{ can be decomposed into two parts denoted as } \hat{H}_{s, \alpha}^{(+)Fr} \text{ and } \hat{H}_{s, \alpha}^{(+)nFr};
\]
\[
\hat{H}_{s, \alpha}^{(+)Fr}(z, \omega_s) = \hat{H}_{s, \alpha}^{(+)Fr}(z, \omega_s) + \hat{H}_{s, \alpha}^{(+)nFr}(z, \omega_s), \quad (5)
\]
\[
\hat{H}_{s, \alpha}^{(+)nFr}(z, \omega_s) = \frac{\kappa_{s, \alpha}^s(\omega_s)}{\mu_0} \hat{E}_{s, \alpha}^{(+)}(z, \omega_s), \quad (6)
\]
\[
\hat{H}_{s, \alpha}^{(+)nFr}(z, \omega_s) = \sqrt{\frac{\hbar c n_s(\omega_s)}{2\mu_0 \omega_s}} \hat{F}_{s, \alpha}(z, \omega_s) \sum_{\beta, \gamma = F, B} \int d\omega | g(\omega_s, \omega_i) \times E_{\beta}^{(+)}(\omega_s + \omega_i) \exp[ik_{0, \beta}(\omega_s + \omega_i)z] \times \exp[-ik_{s, \alpha}(\omega_i)z]a_{\beta}^\dagger(0, \omega_i), \quad \alpha = F, B. \quad (7)
\]

By definition, the magnetic-field amplitude operator \(\hat{H}_{s, \alpha}^{(+)Fr}(z, \omega_s)\) is linearly proportional to the electric-field amplitude operator \(\hat{E}_{s, \alpha}^{(+)}(z, \omega_s)\). The remaining magnetic-field operator \(\hat{H}_{s, \alpha}^{(+)nFr}\) is of purely nonlinear origin and the usual derivation of Fresnel relations does not take it into account. Standard approaches to nonlinear interactions thus do not involve this nonlinear term and so they neglect surface effects. We note that the ‘nonlinear’ magnetic-field operator \(\hat{H}_{s, \alpha}^{(+)nFr}\) occurs as a classical field amplitude also in the description of stimulated parametric processes (e.g., in difference-frequency generation) and yields surface contributions to these processes.

The electric- and magnetic-field amplitudes \(E_{s, \alpha}(z, \omega_m)\) and \(H_{s, \alpha}(z, \omega_m)\) originating in the nonlinear interaction and written in Eqs. (14) and (15) have to obey continuity requirements at the input and output boundaries of the nonlinear medium. We illustrate our approach to this problem considering the signal field at the input boundary (\(z = 0\)). Four electric and magnetic fields are involved in the continuity requirements at this boundary (see Fig. 1): two at the linear left-hand side [denoted by superscript (0)] and two at the nonlinear right-hand side. Because the magnetic-field amplitudes \(H_{s, F}\) and \(H_{s, B}\) inside the nonlinear medium have also nonlinear contributions \(H_{s, F}^{nFr}\) and \(H_{s, B}^{nFr}\) given in Eq. (7) additional (surface) amplitude corrections \(\delta E_{s, F,B}\) and \(\delta E_{s, B}^{(0)}\) [together with \(\delta H_{s,F}^{(0)}\) and \(\delta H_{s,B}^{(0)}\)] in the fields leaving the boundary naturally occur. The amplitude corrections \(\delta E_{s, F,B}^{(0)}\) and \(\delta H_{s,F}^{(0)}\) of the outgoing field outside the nonlinear medium can be involved in the fields obeying Fresnel relations \(\hat{F}\) at the expense of introduction of fictitious amplitude corrections \(\delta E_{s, F,B}\) and \(\delta H_{s,F}\) of the field impinging at the boundary from its nonlinear side. A detailed analysis then results in two equations for the surface amplitude corrections of fields inside the nonlinear medium:
\[
0 = \delta E_{s, F}^{(0)}(0) - \delta E_{s, B}^{(0)}(0),
\]
\[
0 = H_{s, F}^{nFr}(0) + \delta H_{s, F}(0) + H_{s, B}^{nFr}(0) - \delta H_{s, B}(0). \quad (8)
\]

The positive-frequency parts of surface amplitude-correction operators \(\delta \hat{E}_{s, \alpha}^{(+)Fr}\) and \(\delta \hat{H}_{s, \alpha}^{(+)Fr}\) occurring in the quantum form of Eqs. (5) can be expressed using annihilation-operator corrections \(\delta \hat{a}_{s, \alpha}\) similarly to the corresponding amplitude operators \(\hat{E}_{s, \alpha}^{(+)Fr}\) and \(\hat{H}_{s, \alpha}^{(+)Fr}\) in Eqs. (2) and (4). The solution of Eqs. (5) for \(\delta \hat{a}_{s, \alpha}\) and \(\delta \hat{a}_{s, B}\) then takes the form:
\[
\delta \hat{a}_{s, r}(0, \omega_s) = \delta \hat{a}_{s, r}(0, \omega_s) - \frac{i}{\kappa_{s, \alpha}^s(\omega_s)} \sum_{\beta, \gamma = F, B} \int d\omega \, g(\omega_s, \omega_i) E_{\beta}^{(+)}(0, \omega_s + \omega_i) a_{\beta}^\dagger(0, \omega_i). \quad (9)
\]

Similar considerations appropriate for the output boundary leaves us finally with an expression for operators \(\hat{a}_{s, \alpha}(L, \omega_s)\) valid up to the first power of \(g\) (\(L\) stands for the length of nonlinear medium):
\[
\hat{a}_{s, \alpha}(L, \omega_s) = \hat{a}_{s, \alpha}^{\text{free}}(L, \omega_s) + \sum_{\beta, \gamma = F, B} \int d\omega \, \mathcal{F}_{s, \alpha, \beta, \gamma}(\omega_s, \omega_i) a_{\beta}^\dagger(0, \omega_i). \quad (10)
\]

Operators \(\hat{a}_{s, \alpha}^{\text{free}}(L, \omega_s)\) correspond to free-field linear propagation, i.e. without photon-pair generation. The idler-field amplitudes can be analyzed along the same vein.

The generalized two-photon spectral amplitudes \(\mathcal{F}_{s, \alpha, \beta, \gamma}\) and \(\mathcal{F}\) defined in Eq. (10) describe properties of a generated photon pair and are composed of two contributions:
\[
\mathcal{F}_{s, \alpha, \beta, \gamma} = \mathcal{F}_{s, \alpha, \beta, \gamma}^{\text{vol}} + \mathcal{F}_{s, \alpha, \beta, \gamma}^{\text{surf}}; \quad \alpha, \beta, \gamma = F, B. \quad (11)
\]
Two-photon spectral amplitude $\mathcal{F}^{\text{vol}}$ of the volume contribution has the well-known form:

$$
\Phi_{\alpha,\beta,\gamma}^{\text{vol}}(\omega_s, \omega_i) = g(\omega_s, \omega_i)E^{(+)\dagger}(0, \omega_s + \omega_i) \\
\times \exp[ik_{\text{pa}}(\omega_s + \omega_i)L]\exp[-i\Delta k_{\alpha,\beta,\gamma}(\omega_s, \omega_i)L/2] \\
\times L \text{sinc}[\Delta k_{\alpha,\beta,\gamma}(\omega_s, \omega_i)L/2]; \quad \alpha, \beta, \gamma = F, B.
$$

(12)

On the other hand, surface contributions $\mathcal{F}^{m, \text{surf}}$ to the two-photon spectral amplitudes can be expressed as:

$$
\mathcal{F}_{\alpha,\beta,\gamma}^{m, \text{surf}}(\omega_s, \omega_i) = \mathcal{V}_{m, \beta, \gamma}(\omega_s, \omega_i)\Phi_{\alpha,\beta,\gamma}^{\text{vol}}(\omega_s, \omega_i),
$$

(13)

where

$$
\mathcal{V}_{m, \beta, \gamma}(\omega_s, \omega_i) = \frac{\Delta k_{\alpha,\beta,\gamma}(\omega_s, \omega_i)}{k_m(\omega_m)}; \quad \alpha, \beta, \gamma = F, B.
$$

(14)

The structure of surface contributions as described by the two-photon amplitudes $\mathcal{F}^{s, \text{surf}}$ and $\mathcal{F}^{i, \text{surf}}$ resembles that of the volume contribution as the formula in Eq. (13) indicates. The physical interpretation is as follows. At a boundary, the only restriction for photon-pair generation is imposed by the conservation of energy. However, the mutual interference of two-photon amplitudes originating at the input and output boundaries leads to the result that resembles the usual phase-matching conditions. We note that $\lim_{L \to 0} \mathcal{F}^{m, \text{surf}} = 0$.

We further consider photon pairs with both photons propagating forward and use operators $\hat{a}_m(\omega_m) \quad (m = s, i)$ defined outside the nonlinear medium. The joint signal-idler photon-number density $n(\omega_s, \omega_i)$ at the output plane of the nonlinear medium is given as:

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

(15)

Symbol $\langle \rangle$ denotes averaging over the initial signal- and idler-field vacuum state. Introducing two-photon spectral amplitudes $\mathcal{F}^{s}$ and $\mathcal{F}^{i}$ (transmission coefficients $t_m$ describe the output boundary),

$$
\mathcal{F}^{m}(\omega_s, \omega_i) = t_s(\omega_s)t_i(\omega_i)\mathcal{F}_{F,F,F}^{m}(\omega_s, \omega_i), \quad m = s, i,
$$

(16)

we arrive at the following formula:

$$
n(\omega_s, \omega_i) = \text{Re}\{\mathcal{F}^{s*}(\omega_s, \omega_i)\mathcal{F}^{i}(\omega_s, \omega_i)\}.
$$

(17)

As this example illustrates, a generalization of the usual formalism based on a two-photon spectral amplitude can be given providing formulas for all physical quantities characterizing photon pairs.

The volume interaction among the forward-propagating pump, signal, and idler fields dominates in bulk nonlinear crystals several mm long. According to our model, the surface contributions can be approximately included into the usual formalism working with a two-photon spectral amplitude $\Phi^{\text{vol}}$ (see, e.g., [9, 10]) using the formal substitution:

$$
\Phi(\omega_s, \omega_i) \to \sqrt{1 + \mathcal{V}_{F,F,F}^{s}(\omega_s, \omega_i)} \\
\times \sqrt{1 + \mathcal{V}_{F,F,F}^{i}(\omega_s, \omega_i)} \Phi^{\text{vol}}(\omega_s, \omega_i);
$$

(18)

FIG. 2: Signal-field spectra $S_{s}^{\text{vol}}$ (solid curve denoted as a) and $S_{s}^{\text{surf}}$ (solid curve denoted as b) of volume and surface SPDC, respectively, and ratio $S_{s}^{\text{vol}+\text{surf}}/S_{s}^{\text{vol}}$ of the spectra with $S_{s}^{\text{vol}+\text{surf}}$ and without $S_{s}^{\text{vol}}$. The inclusion of surface SPDC gives efficient photon-pair generation (see Fig. 2). Different contributions to surface SPDC from an individual layer is weak but both of them are highly enhanced by constructive interference of fields from different layers. A generalization of the presented theory to layered structures is straightforward following the work presented in [11, 12].

As an example, we consider a structure composed of 25 layers of nonlinear GaN 117 nm thick that sandwich 24 linear layers of AlN 180 nm thick and studied previously in [11]. The volume SPDC gives efficient photon-pair generation at degenerate signal- and idler-field frequencies for the signal-field emission angle 14 deg [11] (see Fig. 2) assuming a normally incident pump field at $\lambda_p = 664.5$ nm and s-polarized fields. Additional photon pairs originate in surface SPDC. Their intensity is cca 20 % of that coming from the volume. However, both contributions are in phase and add constructively so that the inclusion of surface SPDC roughly doubles the number of emitted photon pairs in the spectral area of efficient photon-pair generation (see Fig. 2). Different contributions to surface SPDC can be quantified using coefficients $\mathcal{V}_{F,F,F}^{s}$, defined in Eq. (14). When the lengths of nonlinear layers are less or comparable to the coherence length of the nonlinear process, we observe appreciable contributions of surface terms. For example, the coherence length equals approximately 1 $\mu$m for GaN in our case.

Surface effects give also an important contribution to photon-pair generation rates in periodically-poled nonlinear materials with sufficiently short poling periods.
Surface SPDC is by no means restricted to 1D nonlinear structures: even greater relative contributions are expected in 2D and 3D nonlinear samples. Surface effects will also affect stimulated processes like second-harmonic or second-subharmonic generation when studied under comparable conditions. Qualitatively, they will effectively enhance the nonlinearity. This may be particularly interesting for nonlinear photonic-band-gap fibers.

In conclusion, surface SPDC has been predicted. Generalized signal- and idler-field two-photon spectral amplitudes have been suggested to determine properties of emitted photon pairs. Surface SPDC is important whenever strongly phase-mismatched nonlinear interactions give considerable contributions. This occurs, e.g., in nonlinear layered structures or periodically-poled materials where surface and volume contributions can be comparable. Surface SPDC may affect optimum design of these structures that are considered as promising versatile sources of photon pairs for optoelectronics.

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Here, as an example, we consider frequency-degenerate SPDC in periodically-poled LiNbO$_3$ with the optical axis perpendicular to the direction of collinearly-propagating fields; their polarizations are parallel to the optical axis. Whereas the surface effects contribute to photon-pair generation rate $N_{\text{vol+surf}}$ only by several percent for the pump wavelength $\lambda_p^0 = 1 \, \mu$m, the increase of photon-pair generation rate $N$ by 50% is observed for $\lambda_p^0 = 0.35 \, \mu$m (see Fig. 3). As the curves in Fig. 3 indicate the relative contribution $N_{\text{vol+surf}}/N_{\text{vol}} - 1$ of surface terms is roughly proportional to the inverse $1/\Lambda_{\text{nl}}$ of poling period that is linearly proportional to the density of surfaces per a unit length. We note that domains shorter than 1 $\mu$m can be fabricated using light-induced domain engineering [14].