1 Difference Frequency Generation

This derivation is based on a method developed for birefringently phase matched second harmonic generation [1]. It is adapted to the problem of quasi phase matched difference frequency generation. To include the frequency dependence of the index of refraction to all orders, the difference frequency generation in the PPLN crystal is modeled in the frequency domain. Therefore, Maxwell's propagation equations for the three fields are converted from the time to the frequency domain. The equations can be separated within the reasonable assumption that the Fourier spectra of the pulses at the three frequencies do not overlap.

Some of the equation manipulations and substitutions detailed below have numerical rather than physical reasons. For numerical convenience, it is desirable to shift all spectra to zero frequency, and deal only with the spectral components that are covered by the pulses. Since these spectra represent pulses propagating at the group velocity of the respective pulses, in the laboratory frame, the phase factor of each Fourier component will take very large values with increasing distance z, making numerical computation unnecessary challenging. It is therefore desirable to subtract any giant constant phase factor (which has no physical significance) as well as choose a retarded frame of reference, propagating at the group velocity of one of the pulses. Since the pump pulse repetition rate is the primary clock of this system, it is natural to chose a frame of reference for the three pulses moving at the pump group velocity.

The Fourier transform of Maxwell's wave equation can be written [2] as:

$$\left[\frac{\partial^2}{\partial z^2} + \frac{\Omega^2}{c^2}\epsilon_r(\Omega)\right]\tilde{\mathbf{E}}(\Omega, z) = -\mu_0\Omega^2\tilde{\mathbf{P}}^{NL}(\Omega, z) \tag{1}$$

where

$$\epsilon_r(\Omega) = 1 + \chi^{(1)}(\Omega). \tag{2}$$

is the relative dielectric constant, and $\tilde{\mathbf{E}}(\Omega, z)$ is the total electric field, which includes the pump field centered at ω_p, the signal field centered at ω_s and the idler centered at ω_i. The three frequencies are related by

$$\omega_p = \omega_s + \omega_i. \tag{3}$$

In the nonlinear part of the polarization $\tilde{\mathbf{P}}^{NL}$ we consider only the $\chi^{(2)}$ terms that are associated with the three interacting waves. Each of these terms has to fulfill the wave equation separately. If we also assume that the second order susceptibility $\chi^{(2)}$ does not depend on frequency, the nonlinear polarizations can be decomposed in three contributions, respectively centered at ω_p, ω_s and ω_i to yield for the nonlinear polarizations in a uniform medium:

$$\begin{aligned}
\tilde{\mathbf{P}}_p^{NL}(t,z) &= 2\epsilon_0\chi^{(2)}\tilde{E}_s(t,z)\tilde{E}_i(t,z) \\
\tilde{\mathbf{P}}_s^{NL}(t,z) &= 2\epsilon_0\chi^{(2)}\tilde{E}_p(t,z)\tilde{E}_i^*(t,z) \\
\tilde{\mathbf{P}}_i^{NL}(t,z) &= 2\epsilon_0\chi^{(2)}\tilde{E}_p(t,z)\tilde{E}_s^*(t,z).
\end{aligned} \tag{4}$$

The crystal considered, however is not uniform. It has a domain reversal grating for quasi phase matching. As a consequence $\chi^{(2)}$ reverses sign after a propagation through the crystal of one half times the grating period g ($g \approx 30\mu$m). It is common practice to approximate this sudden domain reversal by a sinusoidal modulation of $\chi^{(2)}$ so that

$$\chi^{(2)} \rightarrow \chi^{(2)}e^{i\Delta kz}. \tag{5}$$

$\Delta k = 2\pi/g$ is the k-vector of the quasi phase matching grating with grating constant g. A more accurate treatment can be done by developing the exact square modulation function into a Fourier series [3]. This treatment reveals, that the approximation of sinusoidal modulation leads to an overestimation of the strength of the interaction by a constant factor of $2/\pi$. With these modifications we can write for Equations (4) in the frequency domain

$$\tilde{\mathbf{P}}_p^{NL}(\Omega, z) = \frac{\chi^{(2)}\epsilon_0}{\pi}\int \tilde{E}_s(\Omega', z)\tilde{E}_i(\Omega - \Omega', z)e^{i\Delta kz}d\Omega'$$

$$\tilde{\mathbf{P}}_s^{NL}(\Omega,z) = \frac{\chi^{(2)}\epsilon_0}{\pi}\int \tilde{E}_i^*(\Omega',z)\tilde{E}_p(\Omega+\Omega',z)e^{-i\Delta kz}d\Omega'$$
$$\tilde{\mathbf{P}}_i^{NL}(\Omega,z) = \frac{\chi^{(2)}\epsilon_0}{\pi}\int \tilde{E}_s^*(\Omega',z)\tilde{E}_p(\Omega+\Omega',z)e^{-i\Delta kz}d\Omega'. \tag{6}$$

Inserting these three polarizations into the wave Eq. (1), and grouping terms of the same central frequency, leads to the set of three equations:

$$\left(\frac{\partial^2}{\partial z^2} + \frac{\Omega^2}{c^2}\epsilon_r(\Omega)\right)E_p(\Omega,z) = -\frac{\epsilon_0\mu_0\Omega^2\chi^{(2)}}{\pi}\int E_s(\Omega',z)E_i(\Omega-\Omega',z)e^{i\Delta kz}d\Omega'$$
$$\left(\frac{\partial^2}{\partial z^2} + \frac{\Omega^2}{c^2}\epsilon_r(\Omega)\right)E_s(\Omega,z) = -\frac{\epsilon_0\mu_0\Omega^2\chi^{(2)}}{\pi}\int E_i^*(\Omega',z)E_p(\Omega+\Omega',z)e^{-i\Delta kz}d\Omega'$$
$$\left(\frac{\partial^2}{\partial z^2} + \frac{\Omega^2}{c^2}\epsilon_r(\Omega)\right)E_i(\Omega,z) = -\frac{\epsilon_0\mu_0\Omega^2\chi^{(2)}}{\pi}\int E_s^*(\Omega',z)E_p(\Omega+\Omega',z)e^{-i\Delta kz}d\Omega'. \tag{7}$$

A description in complex spectral amplitude $\tilde{a}_{p,s,i}(\Omega,z)$ and fast varying spectral phase $-ik(\Omega)z$ is next chosen for each pulse:

$$\tilde{E}_{p,s,i}(\Omega,z) = \frac{1}{2}\tilde{a}_{p,s,i}(\Omega,z)e^{-ik(\Omega)z} \tag{8}$$

Substituting this in Equations (7) and using the condition

$$k^2(\Omega) = \frac{\Omega^2}{c^2}\epsilon_r(\Omega). \tag{9}$$

imposed by Maxwell's wave equation, leads to a set of three differential equations for the evolution with distance of the three complex spectral amplitude functions:

$$\frac{\partial}{\partial z}\tilde{a}_p(\Omega,z) = \frac{-i\Omega^2\chi^{(2)}}{4\pi c^2 k(\Omega)}\int \tilde{a}_s(\Omega',z)\tilde{a}_i(\Omega-\Omega',z)e^{i\left[-k(\Omega')-k(\Omega-\Omega')+k(\Omega)+\Delta k\right]z}d\Omega' - \frac{i}{2k(\Omega)}\frac{\partial^2}{\partial z^2}\tilde{a}_p(\Omega,z)$$
$$\frac{\partial}{\partial z}\tilde{a}_s(\Omega,z) = \frac{-i\Omega^2\chi^{(2)}}{4\pi c^2 k(\Omega)}\int \tilde{a}_i^*(\Omega',z)\tilde{a}_p(\Omega'+\Omega,z)e^{i\left[k(\Omega')-k(\Omega'+\Omega)+k(\Omega)-\Delta k\right]z}d\Omega' - \frac{i}{2k(\Omega)}\frac{\partial^2}{\partial z^2}\tilde{a}_s(\Omega,z)$$
$$\frac{\partial}{\partial z}\tilde{a}_i(\Omega,z) = \frac{-i\Omega^2\chi^{(2)}}{4\pi c^2 k(\Omega)}\int \tilde{a}_s^*(\Omega',z)\tilde{a}_p(\Omega'+\Omega,z)e^{i\left[k(\Omega')-k(\Omega'+\Omega)+k(\Omega)-\Delta k\right]z}d\Omega' - \frac{i}{2k(\Omega)}\frac{\partial^2}{\partial z^2}\tilde{a}_i(\Omega,z). \tag{10}$$

It has been shown [1] that the second derivative is generally negligible, consistent with the slowly varying envelope approximation, even down to a few optical cycles. For convenience of numerical computation, the various spectral envelopes should be centered at the origin of the frequency axis, which is achieved by defining the shifted functions

$$\tilde{\mathcal{E}}_{p,s,i}(\Omega,z) = \tilde{a}_{p,s,i}(\Omega+\omega_{p,s,i},z)$$
$$k_{p,s,i}(\Omega) = k(\Omega+\omega_{p,s,i}) \tag{11}$$
$$\Omega_{p,s,i} = \Omega-\omega_{p,s,i}. \tag{12}$$

In the set of equations that follows, the frequency argument takes symmetric values with respect to the origin, positive and negative, over a range of a few inverse pulse durations.

$$\frac{\partial\tilde{\mathcal{E}}_p(\Omega_p)}{\partial z} = \frac{-i\omega_p^2\chi^{(2)}}{4\pi c^2 k_p(\Omega_p)}\int_{-\infty}^{\infty}\tilde{\mathcal{E}}_s(\Omega_s')\tilde{\mathcal{E}}_i(\Omega_p-\Omega_s')e^{i(-k_s(\Omega_s')-k_i(\Omega_p-\Omega_s')+k_p(\Omega_p)+\Delta k)z}d\Omega_s'$$
$$\frac{\partial\tilde{\mathcal{E}}_s(\Omega_s)}{\partial z} = \frac{-i\omega_s^2\chi^{(2)}}{4\pi c^2 k_s(\Omega_s)}\int_{-\infty}^{\infty}\tilde{\mathcal{E}}_i^*(\Omega_i')\tilde{\mathcal{E}}_p(\Omega_i'+\Omega_s)e^{i(k_i(\Omega_i')-k_p(\Omega_i'+\Omega_s)+k_s(\Omega_s)-\Delta k)z}d\Omega_i'$$
$$\frac{\partial\tilde{\mathcal{E}}_i(\Omega_i)}{\partial z} = \frac{-i\omega_i^2\chi^{(2)}}{4\pi c^2 k_i(\Omega_i)}\int_{-\infty}^{\infty}\tilde{\mathcal{E}}_s^*(\Omega_s')\tilde{\mathcal{E}}_p(\Omega_s'+\Omega_i)e^{i(k_s(\Omega_s')-k_p(\Omega_s'+\Omega_i)+k_i(\Omega_i)-\Delta k)z}d\Omega_s' \tag{13}$$

Here the condition (3) has been used to convert, for example:

$$\begin{aligned}\tilde{a}_i(\Omega-\Omega',z) &= \tilde{\mathcal{E}}_i(\Omega-\Omega'-\omega_i)\\ &= \tilde{\mathcal{E}}_i(\Omega_p+\omega_p-\Omega'_s-\omega_s-\omega_i)\\ &= \tilde{\mathcal{E}}_i(\Omega_p-\Omega'_s).\end{aligned}\tag{14}$$

In order to simplify notation and numerical treatment we can introduce the quantities

$$k'_{p,s,i}(\Omega_{p,s,i}) = k_{p,s,i}(\Omega_{p,s,i}) - k_{p,s,i}(0)\tag{15}$$

to separate the constant k-vectors at the pulses center frequencies from the remaining k dependence that varies throughout each pulse. The quasi phase matching grating responsible for Δk is assumed to have a grating constant g that ensures phase matching at the center frequencies, such that

$$k_p(0) - k_s(0) - k_i(0) + \Delta k = 0.\tag{16}$$

To condense the notation, the following quantities are introduced:

$$\begin{aligned}A_p &= \frac{-i\omega_p^2\chi^{(2)}}{4\pi c^2 k_p(\Omega_p)}\\ A_s &= \frac{-i\omega_s^2\chi^{(2)}}{4\pi c^2 k_s(\Omega_s)}\\ A_i &= \frac{-i\omega_i^2\chi^{(2)}}{4\pi c^2 k_i(\Omega_i)}\end{aligned}\tag{17}$$

$$\begin{aligned}\mathcal{S}(\Omega_s) &= \tilde{\mathcal{E}}_s(\Omega_s)e^{-ik'_s(\Omega_s)z}\\ \mathcal{I}(\Omega_i) &= \tilde{\mathcal{E}}_i(\Omega_i)e^{-ik'_i(\Omega_i)z}\\ \mathcal{P}(\Omega_p) &= \tilde{\mathcal{E}}_p(\Omega_p)e^{-ik'_p(\Omega_p)z}\end{aligned}\tag{18}$$

to obtain the coupled equations for the field amplitudes from Equations (13):

$$\begin{aligned}\frac{\partial\tilde{\mathcal{E}}_p(\Omega_p)}{\partial z} &= A_p\left(\mathcal{S}*\mathcal{I}\right)(\Omega_p)e^{ik'_p(\Omega_p)z}\\ \frac{\partial\tilde{\mathcal{E}}_s(\Omega_s)}{\partial z} &= A_s\left(\mathcal{I}\star\mathcal{P}\right)(\Omega_s)e^{ik'_s(\Omega_s)z}\\ \frac{\partial\tilde{\mathcal{E}}_i(\Omega_i)}{\partial z} &= A_i\left(\mathcal{S}\star\mathcal{P}\right)(\Omega_i)e^{ik'_i(\Omega_i)z}.\end{aligned}\tag{19}$$

Here the operator $*$ describes a convolution and $\star$ a cross-correlation. The indices $n(\Omega_{p,s,i})$ are required to calculate $k_{p,s,i}$. They are obtained from a Sellmeier equation [4].

The change of electric field for each pulse due to the 3 wave interaction is obtained by integrating the set of Equations (19) over the thickness of the OPO crystal, given a set of initial fields at $z = z_0$. The solution of this integration however does not include the effect of the linear dispersion of the crystal on the phase of each individual pulse, because the transformation (8) has removed the effect of dispersion. This transformation has to be reversed after each integration step in order to get the complete electric field. For propagation from z_0 to z_1 the reverse transformation is a multiplication by $\exp[ik'_j(\Omega_j)(z_1-z_0)]$ where j takes the value p, s or i. It is in addition desirable to use a frame of reference moving with the group velocity of the pump pulse:

$$\begin{aligned}\tilde{E}_p(\Omega_p,z_1) &= \tilde{\mathcal{E}}_p(\Omega_p,z_0)e^{i(k'_p(\Omega_p)-\frac{dk'}{d\Omega}|_{\Omega_p}\frac{\Omega_p}{2})(z_1-z_0)}\\ \tilde{E}_s(\Omega_s,z_1) &= \tilde{\mathcal{E}}_s(\Omega_s,z_0)e^{i(k'_s(\Omega_s)-\frac{dk'}{d\Omega}|_{\Omega_p}\frac{\Omega_p}{2})(z_1-z_0)}\\ \tilde{E}_i(\Omega_i,z_1) &= \tilde{\mathcal{E}}_i(\Omega_i,z_0)e^{i(k'_i(\Omega_i)-\frac{dk'}{d\Omega}|_{\Omega_p}\frac{\Omega_p}{2})(z_1-z_0)}.\end{aligned}\tag{20}$$

The first order term in k that is subtracted in each term above corresponds indeed to a displacement in time due to the lower group velocity in the crystal. Since it is common to all three pulses it is not relevant here.

References

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