



## PULSE PROPAGATION IN CHIRAL OPTICAL PARAMETRIC PROCESSES

### Conventions for the complex-valued fields

Throughout this analysis, we stick to the convention that the real-valued electric field  $\mathbf{E}(\mathbf{r}, t)$  and polarization density  $\mathbf{P}(\mathbf{r}, t) = \mathbf{P}^{(L)}(\mathbf{r}, t) + \mathbf{P}^{(NL)}(\mathbf{r}, t)$  are defined in terms of the slowly-varying complex-valued electric field envelope  $\mathbf{E}_\omega(\mathbf{r}, t)$  and polarization density envelope  $\mathbf{P}_\omega(\mathbf{r}, t)$  as

$$\mathbf{E}(\mathbf{r}, t) = \sum_{\omega_\sigma} \text{Re}[\mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) \exp(-i\omega_\sigma t)], \quad (1a)$$

$$\begin{aligned} \mathbf{P}(\mathbf{r}, t) &= \sum_{\omega_\sigma} \text{Re}[\mathbf{P}_{\omega_\sigma}(\mathbf{r}, t) \exp(-i\omega_\sigma t)] \\ &= \sum_{\omega_\sigma} \text{Re}[(\mathbf{P}_{\omega_\sigma}^{(L)}(\mathbf{r}, t) + \mathbf{P}_{\omega_\sigma}^{(NL)}(\mathbf{r}, t)) \exp(-i\omega_\sigma t)], \end{aligned} \quad (1b)$$

where the complex-valued envelope  $\mathbf{P}_{\omega_\sigma}^{(L)}(\mathbf{r}, t)$  contains a constitutive description of all terms linear in the electric field strength and  $\mathbf{P}_{\omega_\sigma}^{(NL)}(\mathbf{r}, t)$  all nonlinear terms.

### The linear part of the constitutive relation

The starting point in the analysis is the wave equation for the propagation of electromagnetic waves in chiral media, with the linear part of the electric polarization density given as

$$\mathbf{P}^{(L)}(\mathbf{r}, t) = \epsilon_0 \mathbf{e}_\mu \int_{-\infty}^{\infty} \left( \chi_{\mu\alpha}(-\omega; \omega) + \gamma_{\mu\alpha\beta}(-\omega; \omega) \frac{\partial}{\partial x_\beta} \right) E_\alpha(\mathbf{r}, \omega) \exp(-i\omega t) d\omega.$$

In this standard form,  $\chi_{\mu\alpha}$  is the linear susceptibility tensor due to electric dipolar interactions, while  $\gamma_{\mu\alpha\beta}$  is the gyration tensor due to electric quadrupolar interactions.

### The governing wave equation

With the constitutive relation for the linear polarization density as above, the nonlinear wave equation for the propagation, including the nonlinear part of the electric polarization density  $\mathbf{P}^{(NL)}$  in the complex-valued form

$$\nabla \times \nabla \times \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) - \mathbf{e}_\mu \left( k_{\mu\alpha}^2(\omega_\sigma) + id_{\mu\alpha}(\omega_\sigma) \frac{\partial}{\partial t} - e_{\mu\alpha}(\omega_\sigma) \frac{\partial^2}{\partial t^2} \right) E_{\omega_\sigma}^\alpha(\mathbf{r}, t) = \mu_0 \omega_\sigma^2 \mathbf{P}_{\omega_\sigma}^{(NL)}(\mathbf{r}, t) \quad (2)$$

where the dispersive coefficients are given in their tensor form as

$$\begin{aligned} d_{\mu\alpha} &= d_{\mu\alpha}(\omega_\sigma) \equiv 2k_{\mu\alpha}(\omega_\sigma) \frac{dk_{\mu\alpha}}{d\omega} \Big|_{\omega_\sigma}, & (\text{no sum}) \\ e_{\mu\alpha} &= e_{\mu\alpha}(\omega_\sigma) \equiv k_{\mu\alpha}(\omega_\sigma) \frac{d^2 k_{\mu\alpha}}{d\omega^2} \Big|_{\omega_\sigma}, & (\text{no sum}) \end{aligned}$$

and where the tensor form of the wave vector  $k_{\mu\alpha}(\omega)$  is defined as

$$k_{\mu\alpha}^2(\omega) = \frac{\omega^2}{c^2} \left( \delta_{\mu\alpha} + \chi_{\mu\alpha}(-\omega; \omega) + \gamma_{\mu\alpha\beta}(-\omega; \omega) \frac{\partial}{\partial x_\beta} \right). \quad (3)$$

Here the derivative is to be interpreted as the result of derivation of the electric field, or “the corresponding wave vector  $\beta$ -component of the electric field”.

### Infinite plane wave approximation

For the case of infinite plane waves, we may replace

$$\nabla \times \nabla \times \rightarrow -\nabla^2 \rightarrow -\frac{d^2}{dz^2},$$

reducing Eq. (2) to

$$\mathbf{e}_\mu \frac{\partial^2 E_{\omega_\sigma}^\mu(z, t)}{\partial z^2} + \mathbf{e}_\mu \left( k_{\mu\alpha}^2(\omega_\sigma) + i d_{\mu\alpha}(\omega_\sigma) \frac{\partial}{\partial t} - e_{\mu\alpha}(\omega_\sigma) \frac{\partial^2}{\partial t^2} \right) E_{\omega_\sigma}^\alpha(z, t) = -\mu_0 \omega_\sigma^2 \mathbf{P}_{\omega_\sigma}^{(\text{NL})}(z, t), \quad (4)$$

where we should keep in mind that the linear chirality is included in the definition of  $k_{\mu\alpha}^2$  in Eq. (3), and that repeated indices should be interpreted under the Einstein convention of summation, here over the coordinates  $\mu = x, y, z$ .

As we will see in the following treatment, in Eq. (4) the term with an explicit  $k_{\mu\alpha}^2(\omega_\sigma)$  as coefficient governs the *phase velocity* in the medium in the vicinity of the centre angular frequency  $\omega_\sigma$ , while the term with  $d_{\mu\alpha}(\omega_\sigma)$  as coefficient determines the *group velocity*  $v_g = (\partial k_{\mu\omega}/\partial \omega)^{-1}$ , and finally the term with  $e_{\mu\alpha}(\omega_\sigma)$  as coefficient determines the *group velocity dispersion* in the medium.

### The wave vector and chiral dispersion coefficients

If we assume that the contribution to the dispersion<sup>1</sup> from the gyration tensor  $\gamma_{\mu\alpha\beta}(-\omega; \omega)$  is negligible compared to the dispersion of the linear electric dipolar susceptibility  $\chi_{\mu\alpha}(-\omega; \omega)$ , we may express the coefficients of the linear term of the wave equation in terms of the relative dielectric permittivity from the dipolar interaction,

$$\varepsilon_{\mu\alpha}(\omega) \equiv \delta_{\mu\alpha} + \chi_{\mu\alpha}(-\omega; \omega),$$

as

$$\begin{aligned} k_{\mu\alpha}(\omega) &= \frac{\omega}{c} \left( \varepsilon_{\mu\alpha}(\omega) + \gamma_{\mu\alpha\beta}(-\omega; \omega) \frac{\partial}{\partial x_\beta} \right)^{1/2} \\ &= \frac{\omega}{c} \varepsilon_{\mu\alpha}^{1/2}(\omega) \left( 1 + \frac{\gamma_{\mu\alpha\beta}(-\omega; \omega)}{\varepsilon_{\mu\alpha}(\omega)} \frac{\partial}{\partial x_\beta} \right)^{1/2} \\ &= \{ (1 + \epsilon)^{1/2} \approx 1 + \epsilon/2, \text{ if } \epsilon \ll 1 \} \\ &\approx \frac{\omega}{c} \varepsilon_{\mu\alpha}^{1/2}(\omega) + \frac{1}{2} \frac{\omega}{c} \frac{\gamma_{\mu\alpha\beta}(-\omega; \omega)}{\varepsilon_{\mu\alpha}^{1/2}(\omega)} \frac{\partial}{\partial x_\beta}. \end{aligned} \quad (5)$$

In other words, in the nomenclature here used, we exclude any nonlocal or magnetic dipolar interaction from the concept of the electric permittivity  $\varepsilon_{\mu\alpha}$ , and keep this entirely as a placeholder for electric dipolar interactions. By defining the effective gyration coefficient as

$$g_{\mu\alpha\beta}(\omega) \equiv \frac{\gamma_{\mu\alpha\beta}(-\omega; \omega)}{2\varepsilon_{\mu\alpha}^{1/2}(\omega)}, \quad (\text{no sum}) \quad (6)$$

the wavevector  $k_{\mu\alpha}$  (phase velocity) and derived dispersive coefficients  $d_{\mu\alpha}$  (group velocity) and  $e_{\mu\alpha}$  (group velocity dispersion) may hence be expressed as

$$k_{\mu\alpha}(\omega_\sigma) \approx \frac{\omega_\sigma}{c} \left( \varepsilon_{\mu\alpha}^{1/2}(\omega_\sigma) + g_{\mu\alpha\beta}(\omega_\sigma) \frac{\partial}{\partial x_\beta} \right), \quad (7a)$$

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<sup>1</sup> In other words the dependence of the material parameter with respect to the angular frequency of light around a centre frequency  $\omega_\sigma$ . This dispersion, which is the primary cause for pulse broadening, should here not be confused by the wider concept of dispersion which is the cause for the, say, medium at the idler, signal and pump frequencies ( $\omega_i, \omega_s, \omega_p$ ) of a parametric process to possess different refractive indices, forming the basis of a potential phase mismatch.

$$d_{\mu\alpha}(\omega_\sigma) \equiv 2k_{\mu\alpha}(\omega_\sigma) \frac{dk_{\mu\alpha}(\omega)}{d\omega} \Big|_{\omega_\sigma} \quad (\text{no sum})$$

$$\approx 2 \underbrace{\frac{\omega}{c} \left( \varepsilon_{\mu\alpha}^{1/2}(\omega_\sigma) + g_{\mu\alpha\beta}(\omega_\sigma) \frac{\partial}{\partial x_\beta} \right)}_{k_{\mu\alpha}(\omega_\sigma)} \frac{d}{d\omega} \underbrace{\left( \frac{\omega}{c} \left( \varepsilon_{\mu\alpha}^{1/2}(\omega) + g_{\mu\alpha\beta}(\omega) \frac{\partial}{\partial x_\beta} \right) \right)}_{k_{\mu\alpha}(\omega)} \Big|_{\omega_\sigma}, \quad (\text{no sum}) \quad (7b)$$

$$e_{\mu\alpha}(\omega_\sigma) \equiv k_{\mu\alpha}(\omega_\sigma) \frac{d^2 k_{\mu\alpha}(\omega)}{d\omega^2} \Big|_{\omega_\sigma} \quad (\text{no sum})$$

$$\approx \underbrace{\frac{\omega}{c} \left( \varepsilon_{\mu\alpha}^{1/2}(\omega_\sigma) + g_{\mu\alpha\beta}(\omega_\sigma) \frac{\partial}{\partial x_\beta} \right)}_{k_{\mu\alpha}(\omega_\sigma)} \frac{d^2}{d\omega^2} \underbrace{\left( \frac{\omega}{c} \left( \varepsilon_{\mu\alpha}^{1/2}(\omega) + g_{\mu\alpha\beta}(\omega) \frac{\partial}{\partial x_\beta} \right) \right)}_{k_{\mu\alpha}(\omega)} \Big|_{\omega_\sigma}. \quad (\text{no sum}) \quad (7c)$$

Again, it should be emphasized that in the case of an otherwise isotropic medium, the square root of the relative electric-dipolar part of the permittivity,  $\varepsilon_{\mu\alpha}^{1/2}(\omega)$ , should be interpreted as the regular electric-dipolar part of the refractive index  $n(\omega)$ .<sup>2</sup>

### Choice of medium

By furthermore applying the symmetries of a certain medium to the form of the susceptibilities, we may put the wave equation in a concrete form, considering the previously outlined approximations. By choosing a medium belonging to point-symmetry group 32 (trigonal), the linear polar (local) susceptibility tensor consists of three non-zero elements of which two are independent,

$$\chi_{xx} = \chi_{yy}, \quad \chi_{zz} \quad (8)$$

while the second-order, rank-three polar susceptibility tensor (governing the optical parametric process) consists of ten non-zero elements of which four are independent,

$$\begin{aligned} \chi_{xxx} &= -\chi_{xyy} = -\chi_{yyx} = -\chi_{yxy}, \\ \chi_{xyz} &= -\chi_{yxz}, \quad \chi_{xzy} = -\chi_{yzx}, \quad \chi_{zxy} = -\chi_{zyx}, \end{aligned} \quad (9)$$

In similar, the chiral properties are described by axial (non-local) tensors, for which the linear chiral rank-three tensor  $\gamma_{\mu\alpha\beta}$  governing the gyrotropy consists of exactly the same set of nonzero elements as the all-electric dipolar  $\chi_{\mu\alpha\beta}$ ,

$$\begin{aligned} \gamma_{xxx} &= -\gamma_{xyy} = -\gamma_{yyx} = -\gamma_{yxy}, \\ \gamma_{xyz} &= -\gamma_{yxz}, \quad \gamma_{xzy} = -\gamma_{yzx}, \quad \gamma_{zxy} = -\gamma_{zyx}, \end{aligned} \quad (10)$$

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<sup>2</sup> If we neglect the chiral nature of the dispersion coefficients, that is to say, by ignoring the gyration coefficients  $g_{\mu\alpha\beta}$  inside the derivatives in  $\omega$ , we obtain the simplified coefficients for the group velocity and group velocity dispersion as

$$d_{\mu\alpha}(\omega_\sigma) \approx 2 \frac{\omega_\sigma}{c} \left( \varepsilon_{\mu\alpha}^{1/2}(\omega_\sigma) + g_{\mu\alpha\beta}(\omega_\sigma) \frac{\partial}{\partial x_\beta} \right) \frac{d}{d\omega} \left( \frac{\omega \varepsilon_{\mu\alpha}^{1/2}(\omega)}{c} \right) \Big|_{\omega_\sigma}, \quad (\text{no sum})$$

$$e_{\mu\alpha}(\omega_\sigma) \approx \frac{\omega_\sigma}{c} \left( \varepsilon_{\mu\alpha}^{1/2}(\omega_\sigma) + g_{\mu\alpha\beta}(\omega_\sigma) \frac{\partial}{\partial x_\beta} \right) \frac{d^2}{d\omega^2} \left( \frac{\omega \varepsilon_{\mu\alpha}^{1/2}(\omega)}{c} \right) \Big|_{\omega_\sigma}. \quad (\text{no sum})$$

However tempting this simplification is, we would in this case unfortunately lose any correction to the group velocity and group velocity dispersion from the chirality, and the only way the chirality would manifest itself would be through the phase velocity, hence ignoring any difference in temporal pulse walk-off between, say, LCP and RCP modes. In order to keep generality, we should hence try to keep the gyration coefficients in the analysis as far as possible.

while the rank-four axial susceptibility tensor (governing the chiral contribution to the optical parametric process) consists of 37 non-zero elements of which 14 are independent,

$$\begin{aligned}
\gamma_{xxxx} &= \gamma_{yyyy} \\
\gamma_{xxyy} &= \gamma_{yyxx} \\
\gamma_{xyxy} &= \gamma_{yxyx} \\
&= -\gamma_{xyyx} - \gamma_{yyxx} + \gamma_{yyyy} \\
&= -\gamma_{yxxy} - \gamma_{yyxx} + \gamma_{yyyy} \\
\gamma_{xxyz} &= -\gamma_{yyyz} = \gamma_{xyxz} = \gamma_{yxxz} \\
\gamma_{xxzx} &= -\gamma_{yyzy} = \gamma_{xyzx} = \gamma_{yxzx} \\
\gamma_{xxzz} &= \gamma_{yyzz} \\
\gamma_{zxxy} &= -\gamma_{zyyy} = \gamma_{zxyx} = \gamma_{yzxx} \\
\gamma_{xzzx} &= \gamma_{yzzy} \\
\gamma_{zxzx} &= \gamma_{yzzy} \\
\gamma_{zxxy} &= -\gamma_{zyyy} = \gamma_{zxyx} = \gamma_{yzxx} \\
\gamma_{zxzx} &= \gamma_{zyyz} \\
\gamma_{zxzx} &= \gamma_{zyyz} \\
\gamma_{zxzx} &= \gamma_{zyyz} \\
\gamma_{zzzz} &= \text{indep.}
\end{aligned} \tag{11}$$

Since we here consider the propagation along the  $z$ -axis of infinite plane waves in the  $(x, y)$ -plane, the only tensor elements which here will be of importance in the sets listed above are the electric dipolar components

$$\chi_{xx} = \chi_{yy}, \quad \chi_{xxx} = -\chi_{xyy} = -\chi_{yyx} = -\chi_{yxy}, \tag{12}$$

and the electric quadrupolar, or chiral, components

$$\gamma_{xyz} = -\gamma_{yxz}, \quad \gamma_{xxy} = -\gamma_{yyx} = \gamma_{yxy} = \gamma_{xyx}. \tag{13}$$

In order to sort out things in order, we will in the following make a brief pre-work before compiling the complete linear and nonlinear polarization densities originating from this set of elements of the susceptibility tensors.

#### *Phase velocity terms in the 32 (trigonal) medium*

When applied to the dispersive terms in Eq. (4), given their form provided by Eqs. (5)–(7) and an electric field strength arbitrarily polarized in the  $(x, y)$ -plane, orthogonal to the direction of propagation along the rotational symmetry axis of the 32 (trigonal) medium, this set of susceptibility tensor elements results in

$$\mathbf{e}_\mu k_{\mu\alpha}^2(\omega_\sigma) E_{\omega_\sigma}^\alpha(\mathbf{r}, t) = \frac{\omega^2 n^2(\omega)}{c^2} \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) - \frac{\omega^2}{c^2} \gamma_{xyz}(-\omega; \omega) \nabla \times \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) \tag{14}$$

where  $n(\omega)$  is the regular dipolar part of the refractive index, defined by

$$n(\omega) = (1 + \chi_{xx}(-\omega; \omega))^{1/2}. \tag{15}$$

From the shape of the terms in the expression provided by Eq. (14), which is governing the phase velocity in the medium, we immediately recognize “ $\nabla \times$ ” as the term which for a field propagating along the  $z$ -axis of rotational symmetry of the 32 (trigonal) medium will contribute an additional, chiral part to the regular dipolar refractive index with  $\pm$  sign to the orthogonal LCP/RCP modes, just as expected.

## Group velocity terms in the 32 (trigonal) medium

In similar, for the first dispersive term with  $d_{\mu\alpha}$  as coefficient<sup>3</sup> governing the group velocity, we obtain

$$\begin{aligned}
\mathbf{e}_\mu d_{\mu\alpha}(\omega_\sigma) \frac{\partial E_{\omega_\sigma}^\alpha(\mathbf{r}, t)}{\partial t} &= \mathbf{e}_\mu 2k_{\mu\alpha}(\omega_\sigma) \frac{dk_{\mu\alpha}(\omega)}{d\omega} \Big|_{\omega_\sigma} \frac{\partial E_{\omega_\sigma}^\alpha(\mathbf{r}, t)}{\partial t} \\
&= 2 \frac{\omega}{c} (n(\omega_\sigma) - g_{xyz}(\omega_\sigma) \nabla \times) \frac{d}{d\omega} \left( \frac{\omega}{c} (n(\omega) - g_{xyz}(\omega) \nabla \times) \right) \Big|_{\omega_\sigma} \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} \\
&= 2(k(\omega_\sigma) - g(\omega_\sigma) \nabla \times) (k'(\omega_\sigma) - g'(\omega_\sigma) \nabla \times) \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} \\
&= 2(k(\omega_\sigma) - g(\omega_\sigma) \nabla \times) \left( k'(\omega_\sigma) \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} - g'(\omega_\sigma) \nabla \times \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} \right) \\
&= 2 \left( k(\omega_\sigma) k'(\omega_\sigma) \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} - g(\omega_\sigma) k'(\omega_\sigma) \nabla \times \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} \right. \\
&\quad \left. - k(\omega_\sigma) g'(\omega_\sigma) \nabla \times \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} - g(\omega_\sigma) g'(\omega_\sigma) \nabla \times \nabla \times \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} \right) \\
&\approx 2 \left( k(\omega_\sigma) k'(\omega_\sigma) \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} - (k'(\omega_\sigma) g(\omega_\sigma) + k(\omega_\sigma) g'(\omega_\sigma)) \nabla \times \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} \right) \\
&= 2 \left( \underbrace{k(\omega_\sigma) k'(\omega_\sigma) \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t}}_{\text{"Classic" group velocity}} \underbrace{- \frac{d(k(\omega)g(\omega))}{d\omega} \Big|_{\omega_\sigma} \frac{\partial}{\partial t} \nabla \times \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}_{\text{Chiral modification to the group velocity}} \right), \\
&\quad \quad \quad v_g(\omega) \sim 1/k'(\omega)
\end{aligned} \tag{16}$$

where we in the approximate step dropped the “ $\nabla \times \nabla \times$ ” term, and where we adopted the following short-hand notations, starting with the dipolar contribution  $k(\omega)$  to the magnitude of the wave vector,

$$k(\omega_\sigma) \equiv \frac{\omega_\sigma n(\omega_\sigma)}{c}, \quad k'(\omega_\sigma) \equiv \frac{dk(\omega)}{d\omega} \Big|_{\omega_\sigma}, \quad k''(\omega_\sigma) \equiv \frac{d^2 k(\omega)}{d\omega^2} \Big|_{\omega_\sigma}, \tag{17}$$

and for the gyration coefficient  $g(\omega)$  and its dispersive components,

$$g(\omega_\sigma) \equiv \frac{\omega_\sigma g_{xyz}(\omega_\sigma)}{c} = \frac{\omega_\sigma \gamma_{xyz}(-\omega_\sigma; \omega_\sigma)}{cn(\omega_\sigma)}, \quad g'(\omega_\sigma) \equiv \frac{dg(\omega)}{d\omega} \Big|_{\omega_\sigma}, \quad g''(\omega_\sigma) \equiv \frac{d^2 g(\omega)}{d\omega^2} \Big|_{\omega_\sigma}. \tag{18}$$

To summarize the product  $k(\omega)g(\omega)$  appearing as a derivative in the final line in the derivation of the dispersive terms, this is given as

$$k(\omega)g(\omega) = \frac{\omega^2}{c^2} \gamma_{xyz}(-\omega; \omega),$$

that is to say a purely chiral component without any inclusion of electric dipolar interactions.

Judging from the shape of the final terms in Eq. (16), we may immediately anticipate that in terms of a circularly polarized decomposition, the “ $\nabla \times$ ” term will make a differential contribution to the LCP/RCP components with equal magnitude but with opposite sign, just in the way the gyration term in Eq. (14) with a similar “ $\nabla \times$ ” term will affect the phase velocity.

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<sup>3</sup> Again under the assumption of wave propagation along the  $z$ -axis of rotational symmetry in a medium belonging to the 32 (trigonal) point-symmetry group.

## Group velocity dispersion terms in the 32 (trigonal) medium

For the second dispersive term with  $e_{\mu\alpha}$  as coefficient<sup>4</sup> governing the group velocity dispersion, we obtain

$$\begin{aligned}
\mathbf{e}_\mu e_{\mu\alpha}(\omega_\sigma) \frac{\partial^2 E_{\omega_\sigma}^\alpha(\mathbf{r}, t)}{\partial t^2} &= \mathbf{e}_\mu k_{\mu\alpha}(\omega_\sigma) \frac{d^2 k_{\mu\alpha}(\omega)}{d\omega^2} \Big|_{\omega_\sigma} \frac{\partial^2 E_{\omega_\sigma}^\alpha(\mathbf{r}, t)}{\partial t^2} \\
&= \frac{\omega}{c} (n(\omega_\sigma) - g_{xyz}(\omega_\sigma) \nabla \times) \frac{d^2}{d\omega^2} \left( \frac{\omega}{c} (n(\omega) - g_{xyz}(\omega) \nabla \times) \right) \Big|_{\omega_\sigma} \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} \\
&= (k(\omega_\sigma) - g(\omega_\sigma) \nabla \times) (k''(\omega_\sigma) - g''(\omega_\sigma) \nabla \times) \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} \\
&= (k(\omega_\sigma) - g(\omega_\sigma) \nabla \times) \left( k''(\omega_\sigma) \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} - g''(\omega_\sigma) \nabla \times \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} \right) \\
&= \left( k(\omega_\sigma) k''(\omega_\sigma) \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} - g(\omega_\sigma) k''(\omega_\sigma) \nabla \times \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} \right. \\
&\quad \left. - k(\omega_\sigma) g''(\omega_\sigma) \nabla \times \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} - g(\omega_\sigma) g''(\omega_\sigma) \nabla \times \nabla \times \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} \right) \\
&\approx \left( k(\omega_\sigma) k''(\omega_\sigma) \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} - (k''(\omega_\sigma) g(\omega_\sigma) + k(\omega_\sigma) g''(\omega_\sigma)) \nabla \times \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} \right) \\
&= \underbrace{\left( k(\omega_\sigma) k''(\omega_\sigma) \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} \right)}_{\text{"Classic" group velocity dispersion}} - \underbrace{\left( \frac{d^2(k(\omega)g(\omega))}{d\omega^2} - 2k'(\omega)g'(\omega) \right) \Big|_{\omega_\sigma} \frac{\partial^2}{\partial t^2} \nabla \times \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}_{\text{Chiral modification to the group velocity dispersion}},
\end{aligned} \tag{19}$$

Again, just as in the case of the previous group velocity terms, we may from the shape of the final terms in Eq. (19), immediately anticipate that in terms of a circularly polarized decomposition, the presence of the “ $\nabla \times$ ” term will make a differential contribution to the LCP/RCP components with equal magnitude but with opposite sign.

## The linear part of the polarization density

To recapitulate, the linear part of the polarization density is in Cartesian coordinates expressed in terms of the linear electric dipolar susceptibility tensor  $\chi_{\mu\alpha}$  of Eq. (12) and linear quadrupolar (non-local) susceptibility tensor  $\gamma_{\mu\alpha\beta}$  of Eq. (13) as

$$\begin{aligned}
\varepsilon_0^{-1} \mathbf{P}_\omega^{(L)} &= \mathbf{e}_x \chi_{xx}(-\omega; \omega) E_\omega^x + \mathbf{e}_y \chi_{yy}(-\omega; \omega) E_\omega^y + \mathbf{e}_x \gamma_{xyz}(-\omega; \omega) \frac{\partial E_\omega^y}{\partial z} + \mathbf{e}_y \gamma_{yxz}(-\omega; \omega) \frac{\partial E_\omega^x}{\partial z} \\
&= \chi_{xx}(-\omega; \omega) (\mathbf{e}_x E_\omega^x + \mathbf{e}_y E_\omega^y) + \gamma_{xyz}(-\omega; \omega) \left( \mathbf{e}_x \frac{\partial E_\omega^y}{\partial z} - \mathbf{e}_y \frac{\partial E_\omega^x}{\partial z} \right) \\
&= \mathbf{e}_x \left( \chi_{xx}(-\omega; \omega) E_\omega^x + \gamma_{xyz}(-\omega; \omega) \frac{\partial E_\omega^y}{\partial z} \right) + \mathbf{e}_y \left( \chi_{xx}(-\omega; \omega) E_\omega^y - \gamma_{xyz}(-\omega; \omega) \frac{\partial E_\omega^x}{\partial z} \right).
\end{aligned}$$

As we express this in terms of the formulation of the wave equation in the presence of dispersion

<sup>4</sup> Yet again under the assumption of wave propagation along the  $z$ -axis of rotational symmetry in a medium belonging to the 32 (trigonal) point-symmetry group.

from Eq. (2), we for the phase velocity, group velocity and group velocity dispersion terms have

$$\begin{aligned}
& \mathbf{e}_\mu \left( \underbrace{k_{\mu\alpha}^2(\omega_\sigma) E_{\omega_\sigma}^\alpha(\mathbf{r}, t)}_{\text{See Eq. (14)}} + i \underbrace{d_{\mu\alpha}(\omega_\sigma) \frac{\partial E_{\omega_\sigma}^\alpha(\mathbf{r}, t)}{\partial t}}_{\text{See Eq. (16)}} - \underbrace{e_{\mu\alpha}(\omega_\sigma) \frac{\partial^2 E_{\omega_\sigma}^\alpha(\mathbf{r}, t)}{\partial t^2}}_{\text{See Eq. (19)}} \right) \\
& \approx \underbrace{\frac{\omega^2 n^2(\omega)}{c^2} \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) - \frac{\omega^2}{c^2} \gamma_{xyz}(-\omega; \omega) \nabla \times \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}_{\text{From Eq. (14)}} \\
& + i 2 \underbrace{\left( k(\omega_\sigma) k'(\omega_\sigma) \frac{\partial \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t} - a'(\omega_\sigma) \frac{\partial}{\partial t} \nabla \times \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) \right)}_{\text{From Eq. (16)}} \\
& - \underbrace{\left( k(\omega_\sigma) k''(\omega_\sigma) \frac{\partial^2 \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t)}{\partial t^2} - b''(\omega_\sigma) \frac{\partial^2}{\partial t^2} \nabla \times \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) \right)}_{\text{From Eq. (19)}}, \tag{20}
\end{aligned}$$

where we defined the coefficient  $a'_\sigma(\omega)$ , for the chiral differential deviation of the group velocity of pulses as

$$a'_\sigma \equiv a'(\omega_\sigma) \equiv \left. \frac{d(k(\omega)g(\omega))}{d\omega} \right|_{\omega_\sigma}, \tag{21}$$

and the coefficient  $b''_\sigma(\omega)$ , for the chiral differential broadening of pulses, or equivalently the differential group velocity dispersion, as

$$b''_\sigma \equiv b''(\omega_\sigma) \equiv \left( \frac{d^2(k(\omega)g(\omega))}{d\omega^2} - 2k'(\omega)g'(\omega) \right) \Big|_{\omega_\sigma}. \tag{22}$$

The “prime” and “bis” on the  $a'(\omega_\sigma)$  and  $b''(\omega_\sigma)$  coefficients are not only cosmetic ornaments to match the corresponding  $k'(\omega_\sigma)$  and  $k''(\omega_\sigma)$  coefficients for the first and second order partial derivatives in time, but also since these coefficients in fact *are* first and second order derivatives.

As the terms of Eq. (20) are inserted into the wave equation (2) we hence obtain the vector wave equation for propagation along the  $z$ -axis as<sup>5</sup>

$$\begin{aligned}
& \nabla \times \nabla \times \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) - \left\{ k_\sigma^2 + 2ik_\sigma k'_\sigma \frac{\partial}{\partial t} - k_\sigma k''_\sigma \frac{\partial^2}{\partial t^2} \right\} \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) \\
& + \left\{ \frac{\omega^2}{c^2} \gamma_{xyz}(-\omega; \omega) + 2ia'_\sigma \frac{\partial}{\partial t} - b''_\sigma \frac{\partial^2}{\partial t^2} \right\} \nabla \times \mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) = \mu_0 \omega_\sigma^2 \mathbf{P}_{\omega_\sigma}^{(\text{NL})}(\mathbf{r}, t). \tag{23}
\end{aligned}$$

### Compiling the wave equation for the temporal envelopes

By adopting the short-hand notation for the dipolar part of the magnitude of the wave vector as

$$k_\sigma = k(\omega_\sigma) = \frac{\omega_\sigma n(\omega_\sigma)}{c}, \quad k'_\sigma = k'(\omega_\sigma) = \left. \frac{d}{d\omega} \left( \frac{\omega n(\omega)}{c} \right) \right|_{\omega_\sigma}, \quad k''_\sigma = k''(\omega_\sigma) = \left. \frac{d^2}{d\omega^2} \left( \frac{\omega n(\omega)}{c} \right) \right|_{\omega_\sigma},$$

<sup>5</sup> In this form, one might think that the  $a'(\omega_\sigma)$  and  $b''(\omega_\sigma)$  coefficients seem to each lack a “ $k(\omega_\sigma)$ ” factor when comparing to the other terms of first and second partial derivatives in time. However, keep in mind that the  $\nabla \times$  operating on the electric field will produce exactly this; hence it is perfectly well that the  $a'(\omega_\sigma)$  and  $b''(\omega_\sigma)$  coefficients, which stem from the chiral nature of the medium, lack this.

the wave equation (4) in the vicinity of each centre frequency  $\omega_\sigma$  of the parametric process, for the envelopes  $E_{\omega_\sigma}^\mu(z, t)$  and of the infinite plane waves, expressed component-wise from Eq. (23) for Cartesian coordinates  $\mu = x, y$ , becomes

$$\begin{aligned} \frac{\partial^2 E_{\omega_\sigma}^x(z, t)}{\partial z^2} + \left( k_\sigma^2 + 2ik_\sigma k'_\sigma \frac{\partial}{\partial t} - k_\sigma k''_\sigma \frac{\partial^2}{\partial t^2} \right) E_{\omega_\sigma}^x(z, t) \\ + \left( \frac{\omega_\sigma^2 \gamma_\sigma}{c^2} + 2ia'_\sigma \frac{\partial}{\partial t} - b''_\sigma \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_\sigma}^y(z, t)}{\partial z} = -\mu_0 \omega_\sigma^2 P_{\omega_\sigma}^{(\text{NL})x}(z, t), \end{aligned} \quad (24a)$$

$$\begin{aligned} \frac{\partial^2 E_{\omega_\sigma}^y(z, t)}{\partial z^2} + \left( k_\sigma^2 + 2ik_\sigma k'_\sigma \frac{\partial}{\partial t} - k_\sigma k''_\sigma \frac{\partial^2}{\partial t^2} \right) E_{\omega_\sigma}^y(z, t) \\ - \left( \frac{\omega_\sigma^2 \gamma_\sigma}{c^2} + 2ia'_\sigma \frac{\partial}{\partial t} - b''_\sigma \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_\sigma}^x(z, t)}{\partial z} = -\mu_0 \omega_\sigma^2 P_{\omega_\sigma}^{(\text{NL})y}(z, t), \end{aligned} \quad (24b)$$

where, just to recapitulate the notation, the coefficients  $a'_\sigma \equiv a'(\omega_\sigma)$  and  $b''_\sigma \equiv b''(\omega_\sigma)$  are given by Eqs. (21) and (22), respectively, and where

$$n_\sigma \equiv n(\omega_\sigma) = (1 + \chi_{xx}(-\omega_\sigma; \omega_\sigma))^{1/2}$$

is the dipolar linear part of the regular refractive index, and

$$\gamma_\sigma \equiv \gamma_{xyz}(-\omega_\sigma; \omega_\sigma)$$

the corresponding chiral correction to the refractive index from the non-local (chiral) part of the light-matter interaction.

### The nonlinear polarization density expressed in a Cartesian coordinate system

In the one-dimensional wave equation (24) for the respective centre frequencies  $\omega_\sigma = \omega_i, \omega_s, \omega_p$ , we have yet to explicitly state the nature of the nonlinear wave interactions which act as the mixer between the involved frequencies. From the nonzero applicable sets of susceptibility elements of Eqs. (12) and (13), we obtain the nonlinear polarization density expressed in cartesian coordinates as follows.

#### Nonlinear polarization density for the idler

Given the nonzero tensor elements as listed in Eqs. (12) and (13), we for the idler, at angular frequency  $\omega_1 = \omega_3 - \omega_2$ , have the nonlinear polarization density expressed in the Cartesian coordinate system<sup>6</sup> as

$$\begin{aligned} \varepsilon_0^{-1} \mathbf{P}_{\omega_1}^{(\text{NL})} = & \mathbf{e}_x [\chi_{xxx}(-\omega_1; \omega_3, -\omega_2) E_{\omega_3}^x E_{\omega_2}^{x*} + \chi_{xyy}(-\omega_1; \omega_3, -\omega_2) E_{\omega_3}^y E_{\omega_2}^{y*}] \\ & + \mathbf{e}_y [\chi_{yxy}(-\omega_1; \omega_3, -\omega_2) E_{\omega_3}^x E_{\omega_2}^{y*} + \chi_{yyx}(-\omega_1; \omega_3, -\omega_2) E_{\omega_3}^y E_{\omega_2}^{x*}] \\ & + \mathbf{e}_x [\gamma_{xxyz}(-\omega_1; \omega_3, -\omega_2) \frac{\partial}{\partial z} (E_{\omega_3}^x E_{\omega_2}^{y*}) + \gamma_{xyxz}(-\omega_1; \omega_3, -\omega_2) \frac{\partial}{\partial z} (E_{\omega_3}^y E_{\omega_2}^{x*})] \\ & + \mathbf{e}_y [\gamma_{yyyz}(-\omega_1; \omega_3, -\omega_2) \frac{\partial}{\partial z} (E_{\omega_3}^y E_{\omega_2}^{y*}) + \gamma_{yxxz}(-\omega_1; \omega_3, -\omega_2) \frac{\partial}{\partial z} (E_{\omega_3}^x E_{\omega_2}^{x*})]. \end{aligned}$$

where we for the sake of simplicity in notation omitted the explicit arguments to the temporal field envelopes  $E_{\omega_k}^\mu \equiv E_{\omega_k}^\mu(z, t)$ . By using the relations from Eqs. (12) and (13), which are results of

<sup>6</sup> And, of course, with the susceptibilities expressed with their frequency arguments following the convention of P. N. Butcher and D. Cotter's *The Elements of Nonlinear Optics* (Cambridge, 1990).



spatial symmetry considerations and always hold, regardless of frequency arguments of frequency regime, we may reduce this to the single unique elements  $\chi_{xxx}$  and  $\gamma_{xyz}$  to yield

$$\begin{aligned} \varepsilon_0^{-1} \mathbf{P}_{\omega_1}^{(\text{NL})} = & \chi_{xxx}(-\omega_1; \omega_3, -\omega_2) [\mathbf{e}_x (E_{\omega_3}^x E_{\omega_2}^{x*} - E_{\omega_3}^y E_{\omega_2}^{y*}) - \mathbf{e}_y (E_{\omega_3}^x E_{\omega_2}^{y*} + E_{\omega_3}^y E_{\omega_2}^{x*})] \\ & + \gamma_{xyz}(-\omega_1; \omega_3, -\omega_2) \frac{\partial}{\partial z} [\mathbf{e}_x (E_{\omega_3}^x E_{\omega_2}^{y*} + E_{\omega_3}^y E_{\omega_2}^{x*}) - \mathbf{e}_y (E_{\omega_3}^y E_{\omega_2}^{y*} - E_{\omega_3}^x E_{\omega_2}^{x*})]. \end{aligned} \quad (25)$$

We should here emphasize the complex conjugation of the fields associated with the signal at  $\omega_2$ , connected to the negative frequency argument of  $\omega_2$  in the nonlinear susceptibility tensors  $\chi_{\mu\alpha\beta}(-\omega_1; \omega_3, -\omega_2)$  and  $\gamma_{\mu\alpha\beta\gamma}(-\omega_1; \omega_3, -\omega_2)$ .

#### Nonlinear polarization density for the signal

In the same way as for the idler, we for the signal, at angular frequency  $\omega_2 = \omega_3 - \omega_1$ , obtain the nonlinear polarization density expressed in the Cartesian coordinate system as

$$\begin{aligned} \varepsilon_0^{-1} \mathbf{P}_{\omega_2}^{(\text{NL})} = & \chi_{xxx}(-\omega_2; \omega_3, -\omega_1) [\mathbf{e}_x (E_{\omega_3}^x E_{\omega_1}^{x*} - E_{\omega_3}^y E_{\omega_1}^{y*}) - \mathbf{e}_y (E_{\omega_3}^x E_{\omega_1}^{y*} + E_{\omega_3}^y E_{\omega_1}^{x*})] \\ & + \gamma_{xyz}(-\omega_2; \omega_3, -\omega_1) \frac{\partial}{\partial z} [\mathbf{e}_x (E_{\omega_3}^x E_{\omega_1}^{y*} + E_{\omega_3}^y E_{\omega_1}^{x*}) - \mathbf{e}_y (E_{\omega_3}^y E_{\omega_1}^{y*} - E_{\omega_3}^x E_{\omega_1}^{x*})]. \end{aligned} \quad (26)$$

where the only difference compared to Eq. (25) is the swap of idler and signal angular frequencies  $\omega_1 \rightleftharpoons \omega_2$ . Again, we emphasize the complex conjugation of the fields associated with the idler at  $\omega_1$ , connected to the negative frequency argument of  $\omega_1$  in the nonlinear susceptibility tensors  $\chi_{\mu\alpha\beta}(-\omega_2; \omega_3, -\omega_1)$  and  $\gamma_{\mu\alpha\beta\gamma}(-\omega_2; \omega_3, -\omega_1)$ .

#### Nonlinear polarization density for the pump

In the same way as for the idler and signal, we for the pump, at angular frequency  $\omega_3 = \omega_1 + \omega_2$ , obtain the nonlinear polarization density expressed in the Cartesian coordinate system as

$$\begin{aligned} \varepsilon_0^{-1} \mathbf{P}_{\omega_3}^{(\text{NL})} = & \chi_{xxx}(-\omega_3; \omega_1, \omega_2) [\mathbf{e}_x (E_{\omega_1}^x E_{\omega_2}^x - E_{\omega_1}^y E_{\omega_2}^y) - \mathbf{e}_y (E_{\omega_1}^x E_{\omega_2}^y + E_{\omega_1}^y E_{\omega_2}^x)] \\ & + \gamma_{xyz}(-\omega_3; \omega_1, \omega_2) \frac{\partial}{\partial z} [\mathbf{e}_x (E_{\omega_1}^x E_{\omega_2}^y + E_{\omega_1}^y E_{\omega_2}^x) - \mathbf{e}_y (E_{\omega_1}^y E_{\omega_2}^y - E_{\omega_1}^x E_{\omega_2}^x)]. \end{aligned} \quad (27)$$

In contrary to the nonlinear polarization densities for the idler and signal waves, there is in the corresponding expression for the pump at  $\omega_3 = \omega_1 + \omega_2$  no complex conjugation of the mixed fields, connected to the all-positive frequency arguments of  $\omega_1$  and  $\omega_2$  in the nonlinear susceptibility tensors  $\chi_{\mu\alpha\beta}(-\omega_3; \omega_1, \omega_2)$  and  $\gamma_{\mu\alpha\beta\gamma}(-\omega_3; \omega_1, \omega_2)$ .

#### Inserting the nonlinear polarization density into the wave equation

At this stage, we have all the nonlinear polarization densities for the idler, signal and pump derived, for the electric dipolar part (tensor  $\chi_{\mu\alpha\beta}$ ) as well as the non-local, quadrupolar part (tensor  $\gamma_{\mu\alpha\beta\gamma}$ ). These nonlinear polarization densities will act as sources in the otherwise homogeneous partial differential equations for the temporal field envelopes,<sup>7</sup> and by inserting Eqs. (25)–(27) consecutively into Eq. (24) for the temporal field envelopes, we in Cartesian coordinates obtain the respective wave equation for the total fields of the idler, signal and pump.

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<sup>7</sup> By *temporal field envelope*, we here mean a field where the rapid harmonic oscillation in time has been removed from the equation, by the convention of notation for complex-valued fields as introduced in Eqs. (1),

$$\begin{aligned} \mathbf{E}(\mathbf{r}, t) &= \sum_{\omega_\sigma} \text{Re}[\mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) \exp(-i\omega_\sigma t)], \\ \mathbf{P}(\mathbf{r}, t) &= \sum_{\omega_\sigma} \text{Re}[(\mathbf{P}_{\omega_\sigma}^{(\text{L})}(\mathbf{r}, t) + \mathbf{P}_{\omega_\sigma}^{(\text{NL})}(\mathbf{r}, t)) \exp(-i\omega_\sigma t)]. \end{aligned}$$

It should here be emphasized that we so far have not separated these total fields into any forward and backward traveling components; this will come as soon as we make an ansatz of the temporal envelopes having typical  $\exp(i\omega n_k z/c)$  or  $\exp(-i\omega n_k z/c)$  dependencies, for the forward and backward traveling components, respectively.

*Wave equation for the temporal envelope of the total field of the idler*

To start with, by inserting Eqs. (25) into Eq. (24), we for the temporal field envelope of the idler in Cartesian coordinates obtain

$$\begin{aligned} \frac{\partial^2 E_{\omega_1}^x}{\partial z^2} + \left( k_1^2 + 2ik_1 k_1' \frac{\partial}{\partial t} - k_1 k_1'' \frac{\partial^2}{\partial t^2} \right) E_{\omega_1}^x + \left( \frac{\omega_1^2 \gamma_1}{c^2} + 2ia_1' \frac{\partial}{\partial t} - b_1'' \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_1}^y}{\partial z} \\ = - \left( \frac{\omega_1}{c} \right)^2 \left[ \chi_{xxx}(-\omega_1; \omega_3, -\omega_2) (E_{\omega_3}^x E_{\omega_2}^{x*} - E_{\omega_3}^y E_{\omega_2}^{y*}) \right. \\ \left. + \gamma_{xyz}(-\omega_1; \omega_3, -\omega_2) \frac{\partial}{\partial z} (E_{\omega_3}^x E_{\omega_2}^{y*} + E_{\omega_3}^y E_{\omega_2}^{x*}) \right], \quad (28a) \end{aligned}$$

$$\begin{aligned} \frac{\partial^2 E_{\omega_1}^y}{\partial z^2} + \left( k_1^2 + 2ik_1 k_1' \frac{\partial}{\partial t} - k_1 k_1'' \frac{\partial^2}{\partial t^2} \right) E_{\omega_1}^y - \left( \frac{\omega_1^2 \gamma_1}{c^2} + 2ia_1' \frac{\partial}{\partial t} - b_1'' \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_1}^x}{\partial z} \\ = - \left( \frac{\omega_1}{c} \right)^2 \left[ -\chi_{xxx}(-\omega_1; \omega_3, -\omega_2) (E_{\omega_3}^x E_{\omega_2}^{y*} + E_{\omega_3}^y E_{\omega_2}^{x*}) \right. \\ \left. - \gamma_{xyz}(-\omega_1; \omega_3, -\omega_2) \frac{\partial}{\partial z} (E_{\omega_3}^y E_{\omega_2}^{y*} - E_{\omega_3}^x E_{\omega_2}^{x*}) \right], \quad (28b) \end{aligned}$$

where we used  $\varepsilon_0 \mu_0 \equiv c^{-2}$ , and where we for the sake of simplicity dropped the explicit spatial and temporal arguments of the field envelopes  $E_{\omega_k}^\mu \equiv E_{\omega_k}^\mu(z, t)$ , introduced the short-hand notation

$$n_k \equiv n(\omega_k) \equiv (1 + \chi_{xx}(-\omega_k; \omega_k))^{1/2}, \quad \gamma_k \equiv \gamma(\omega_k) \equiv \gamma_{xyz}(-\omega_k; \omega_k),$$

and, for the coefficients of dispersion,

$$k_k' \equiv k'(\omega_k) \equiv \frac{d}{d\omega} \left( \frac{\omega n(\omega)}{c} \right) \Big|_{\omega_k}, \quad k_k'' \equiv k''(\omega_k) \equiv \frac{d^2}{d\omega^2} \left( \frac{\omega n(\omega)}{c} \right) \Big|_{\omega_k},$$

while the coefficients  $a_\sigma' \equiv a'(\omega_\sigma)$  and  $b_\sigma'' \equiv b''(\omega_\sigma)$  as previously are given by Eqs. (21) and (22), respectively.

*Wave equation for the temporal envelope of the total field of the signal*

Using the same notation as introduced for the idler in the previous section, we by inserting Eqs. (26) into Eq. (24), for the temporal field envelope of the signal, obtain

$$\begin{aligned} \frac{\partial^2 E_{\omega_2}^x}{\partial z^2} + \left( k_2^2 + 2ik_2 k_2' \frac{\partial}{\partial t} - k_2 k_2'' \frac{\partial^2}{\partial t^2} \right) E_{\omega_2}^x + \left( \frac{\omega_2^2 \gamma_2}{c^2} + 2ia_2' \frac{\partial}{\partial t} - b_2'' \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_2}^y}{\partial z} \\ = - \left( \frac{\omega_2}{c} \right)^2 \left[ \chi_{xxx}(-\omega_2; \omega_3, -\omega_1) (E_{\omega_3}^x E_{\omega_1}^{x*} - E_{\omega_3}^y E_{\omega_1}^{y*}) \right. \\ \left. + \gamma_{xyz}(-\omega_2; \omega_3, -\omega_1) \frac{\partial}{\partial z} (E_{\omega_3}^x E_{\omega_1}^{y*} + E_{\omega_3}^y E_{\omega_1}^{x*}) \right], \quad (29a) \end{aligned}$$

$$\begin{aligned} \frac{\partial^2 E_{\omega_2}^y}{\partial z^2} + \left( k_2^2 + 2ik_2 k_2' \frac{\partial}{\partial t} - k_2 k_2'' \frac{\partial^2}{\partial t^2} \right) E_{\omega_2}^y - \left( \frac{\omega_2^2 \gamma_2}{c^2} + 2ia_2' \frac{\partial}{\partial t} - b_2'' \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_2}^x}{\partial z} \\ = - \left( \frac{\omega_2}{c} \right)^2 \left[ -\chi_{xxx}(-\omega_2; \omega_3, -\omega_1) (E_{\omega_3}^x E_{\omega_1}^{y*} + E_{\omega_3}^y E_{\omega_1}^{x*}) \right. \\ \left. - \gamma_{xyz}(-\omega_2; \omega_3, -\omega_1) \frac{\partial}{\partial z} (E_{\omega_3}^y E_{\omega_1}^{y*} - E_{\omega_3}^x E_{\omega_1}^{x*}) \right]. \quad (29b) \end{aligned}$$

Wave equation for the temporal envelope of the total field of the pump

Finally, by inserting Eqs. (27) into Eq. (24), for the temporal field envelope of the pump, we obtain

$$\begin{aligned} \frac{\partial^2 E_{\omega_3}^x}{\partial z^2} + \left( k_3^2 + 2ik_3k_3' \frac{\partial}{\partial t} - k_3k_3'' \frac{\partial^2}{\partial t^2} \right) E_{\omega_3}^x + \left( \frac{\omega_3^2 \gamma_3}{c^2} + 2ia_3' \frac{\partial}{\partial t} - b_3'' \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_3}^y}{\partial z} \\ = - \left( \frac{\omega_3}{c} \right)^2 \left[ \chi_{xxx}(-\omega_3; \omega_1, \omega_2) (E_{\omega_1}^x E_{\omega_2}^x - E_{\omega_1}^y E_{\omega_2}^y) \right. \\ \left. + \gamma_{xyz}(-\omega_3; \omega_1, \omega_2) \frac{\partial}{\partial z} (E_{\omega_1}^x E_{\omega_2}^y + E_{\omega_1}^y E_{\omega_2}^x) \right], \end{aligned} \quad (30a)$$

$$\begin{aligned} \frac{\partial^2 E_{\omega_3}^y}{\partial z^2} + \left( k_3^2 + 2ik_3k_3' \frac{\partial}{\partial t} - k_3k_3'' \frac{\partial^2}{\partial t^2} \right) E_{\omega_3}^y - \left( \frac{\omega_3^2 \gamma_3}{c^2} + 2ia_3' \frac{\partial}{\partial t} - b_3'' \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_3}^x}{\partial z} \\ = - \left( \frac{\omega_3}{c} \right)^2 \left[ -\chi_{xxx}(-\omega_3; \omega_1, \omega_2) (E_{\omega_1}^x E_{\omega_2}^y + E_{\omega_1}^y E_{\omega_2}^x) \right. \\ \left. - \gamma_{xyz}(-\omega_3; \omega_1, \omega_2) \frac{\partial}{\partial z} (E_{\omega_1}^y E_{\omega_2}^y - E_{\omega_1}^x E_{\omega_2}^x) \right]. \end{aligned} \quad (30b)$$

Again, notice the adsence of complex conjugation of mixed fields in the right-hand side of these equations for the tempporal field envelope for the pump.

### Expressing the wave equations in circularly polarized basis vectors

We *á priori* know that the gyrotropic nature introduced by the nonlocal interaction will manifest itself as a circular birefringence. Therefore, we may now carry through the straightforward but nevertheless somewhat cumbersome exercise of expressing the wave equations (28)–(30) in a circularly polarized base.

*The circularly polarized basis vectors*

By using the convention from Eq. (1),

$$\mathbf{E}(\mathbf{r}, t) = \sum_{\omega_\sigma} \text{Re}[\mathbf{E}_{\omega_\sigma}(\mathbf{r}, t) \exp(-i\omega_\sigma t)],$$

for the notation of complex-valued temporal field envelopes, the appropriate sign convention for the circularly polarized base vectors  $\mathbf{e}_+$  (left circular polarization, LCP) and  $\mathbf{e}_-$  (right circular polarization, RCP) yields

$$\mathbf{e}_+ = \frac{1}{\sqrt{2}}(\mathbf{e}_x + i\mathbf{e}_y), \quad \mathbf{e}_- = \frac{1}{\sqrt{2}}(\mathbf{e}_x - i\mathbf{e}_y). \quad (31)$$

The orthogonality properties of these basis vectors yield

$$\mathbf{e}_\pm^* \cdot \mathbf{e}_\pm = 1, \quad \mathbf{e}_\pm^* \cdot \mathbf{e}_\mp = 0, \quad \mathbf{e}_\pm \times \mathbf{e}_\mp = \mp i\mathbf{e}_z, \quad \mathbf{e}_\pm \times \mathbf{e}_z = \pm i\mathbf{e}_\pm. \quad (32)$$

We may equally well inversely express the Cartesian basis vectors  $\mathbf{e}_x$  and  $\mathbf{e}_y$  in terms of the circularly polarized basis vectors as

$$\mathbf{e}_x = \frac{1}{\sqrt{2}}(\mathbf{e}_+ + \mathbf{e}_-), \quad \mathbf{e}_y = \frac{1}{i\sqrt{2}}(\mathbf{e}_+ - \mathbf{e}_-). \quad (33)$$

### The circularly polarized field components

The previously defined circularly polarized basis vectors provide a very convenient tool when it comes to the formulation of the circularly polarized field representation, since we may just project the circularly polarized field components  $E_\omega^+$  (LCP) and  $E_\omega^-$  (RCP) out of the Cartesian representation by

$$E_\omega^+ = \mathbf{e}_+^* \cdot (E_\omega^x \mathbf{e}_x + E_\omega^y \mathbf{e}_y) = \frac{1}{\sqrt{2}}(\mathbf{e}_x - i\mathbf{e}_y) \cdot (E_\omega^x \mathbf{e}_x + E_\omega^y \mathbf{e}_y) = \frac{1}{\sqrt{2}}(E_\omega^x - iE_\omega^y), \quad (34a)$$

$$E_\omega^- = \mathbf{e}_-^* \cdot (E_\omega^x \mathbf{e}_x + E_\omega^y \mathbf{e}_y) = \frac{1}{\sqrt{2}}(\mathbf{e}_x + i\mathbf{e}_y) \cdot (E_\omega^x \mathbf{e}_x + E_\omega^y \mathbf{e}_y) = \frac{1}{\sqrt{2}}(E_\omega^x + iE_\omega^y). \quad (34b)$$

Of course, the projection of a circularly polarized component from a field which already is formulated in the circularly polarized basis is trivial, using the orthogonality conditions  $\mathbf{e}_\pm^* \cdot \mathbf{e}_\pm = 1$  and  $\mathbf{e}_\pm^* \cdot \mathbf{e}_\mp = 0$ ,

$$E_\omega^\pm = \mathbf{e}_\pm^* \cdot (E_\omega^+ \mathbf{e}_+ + E_\omega^- \mathbf{e}_-).$$

Again, we may just as well inverse these relations and express the fields in the Cartesian coordinate system in terms of these circularly polarized components, as

$$E_\omega^x = \frac{1}{\sqrt{2}}(E_\omega^+ + E_\omega^-), \quad E_\omega^y = \frac{i}{\sqrt{2}}(E_\omega^+ - E_\omega^-). \quad (35)$$

Since we in the wave equations (28)–(30) have all fields expressed in a Cartesian system, we will in the formulation of the equivalent systems in a circularly polarized basis primarily use Eqs. (33) and (35).

### The circularly polarized representation of the wave equation for the idler

By expressing all fields in the wave equation (28) for the temporal envelope of the idler at angular frequency  $\omega_1 = \omega_3 - \omega_2$  in the circularly polarized basis from Eq. (33), in terms of the LCP and RCP field components as provided by Eq. (35), we after some cumbersome but straightforward algebra obtain<sup>8</sup>

$$\left[ \frac{\partial^2}{\partial z^2} + k_1^2 + 2ik_1k_1' \frac{\partial}{\partial t} - k_1k_1'' \frac{\partial^2}{\partial t^2} \pm i \left( \frac{\omega_1^2 \gamma_1}{c^2} + 2ia_1' \frac{\partial}{\partial t} - b_1'' \frac{\partial^2}{\partial t^2} \right) \frac{\partial}{\partial z} \right] E_{\omega_1}^\pm = - \left( \frac{\omega_1}{c} \right)^2 \left( p_1 \pm iq_1 \frac{\partial}{\partial z} \right) (E_{\omega_3}^\mp E_{\omega_2}^{\pm*}). \quad (36)$$

with, just to recapitulate, the short-hand notation for the coefficients as

$$\begin{aligned} n_1 &\equiv n(\omega_1) = [1 + \chi_{xx}(-\omega_1; \omega_1)]^{1/2}, & \gamma_1 &\equiv \gamma_{xyz}(-\omega_1; \omega_1), \\ p_1 &\equiv 2^{1/2} \chi_{xxx}(-\omega_1; \omega_3, -\omega_2), & q_1 &\equiv 2^{1/2} \gamma_{xyz}(-\omega_1; \omega_3, -\omega_2), \\ k_1 &\equiv k(\omega_1) \equiv \frac{\omega_1 n(\omega_1)}{c}, & k_1' &\equiv k'(\omega_1) \equiv \frac{d}{d\omega} \left( \frac{\omega n(\omega)}{c} \right) \Big|_{\omega_1}, & k_1'' &\equiv k''(\omega_1) \equiv \frac{d^2}{d\omega^2} \left( \frac{\omega n(\omega)}{c} \right) \Big|_{\omega_1}, \\ a_1' &\equiv a'(\omega_1) \equiv \frac{d(k(\omega)g(\omega))}{d\omega} \Big|_{\omega_1}, & b_1'' &\equiv b''(\omega_1) \equiv \left( \frac{d^2(k(\omega)g(\omega))}{d\omega^2} - 2k'(\omega)g'(\omega) \right) \Big|_{\omega_1}, \\ g(\omega) &\equiv \frac{\omega g_{xyz}(\omega)}{c} = \frac{\omega \gamma_{xyz}(-\omega; \omega)}{cn(\omega)}, & g'(\omega) &\equiv \frac{dg(\omega)}{d\omega}, \end{aligned}$$

where the coefficients  $g(\omega_1)$ ,  $a_1' \equiv a'(\omega_1)$  and  $b_1'' \equiv b''(\omega_1)$  as previously are originally defined by Eqs. (18), (21) and (22), respectively.

<sup>8</sup> From a practical algebraic point, we in the derivation of this expression simply express all Cartesian field components in terms of their circularly polarized components via Eqs. (35) and solve for  $E_{\omega_1}^+$  and  $E_{\omega_1}^-$ .

*The circularly polarized representation of the wave equation for the signal*

In similar to the previous expression for the idler, we by expressing all fields in the wave equation (29) for the temporal envelope of the signal at angular frequency  $\omega_2 = \omega_3 - \omega_1$  in the circularly polarized basis from Eq. (33) obtain

$$\left[ \frac{\partial^2}{\partial z^2} + k_2^2 + 2ik_2k_2' \frac{\partial}{\partial t} - k_2k_2'' \frac{\partial^2}{\partial t^2} \pm i \left( \frac{\omega_2^2 \gamma_2}{c^2} + 2ia_2' \frac{\partial}{\partial t} - b_2'' \frac{\partial^2}{\partial t^2} \right) \frac{\partial}{\partial z} \right] E_{\omega_2}^{\pm} = - \left( \frac{\omega_2}{c} \right)^2 \left( p_2 \pm iq_2 \frac{\partial}{\partial z} \right) (E_{\omega_3}^{\mp} E_{\omega_1}^{\pm*}). \quad (37)$$

with, just to recapitulate, the short-hand notation for the coefficients as

$$\begin{aligned} n_2 &= n(\omega_2) = [1 + \chi_{xx}(-\omega_2; \omega_2)]^{1/2}, & \gamma_2 &= \gamma_{xyz}(-\omega_2; \omega_2), \\ p_2 &= 2^{1/2} \chi_{xxx}(-\omega_2; \omega_3, -\omega_1), & q_2 &= 2^{1/2} \gamma_{xyz}(-\omega_2; \omega_3, -\omega_1), \\ k_2 &\equiv k(\omega_2) \equiv \frac{\omega_2 n(\omega_2)}{c}, & k_2' &\equiv k'(\omega_2) \equiv \frac{d}{d\omega} \left( \frac{\omega n(\omega)}{c} \right) \Big|_{\omega_2}, & k_2'' &\equiv k''(\omega_2) \equiv \frac{d^2}{d\omega^2} \left( \frac{\omega n(\omega)}{c} \right) \Big|_{\omega_2}. \\ a_2' &\equiv a'(\omega_2) \equiv \frac{d(k(\omega)g(\omega))}{d\omega} \Big|_{\omega_2}, & b_2'' &\equiv b''(\omega_2) \equiv \left( \frac{d^2(k(\omega)g(\omega))}{d\omega^2} - 2k'(\omega)g'(\omega) \right) \Big|_{\omega_2}, \end{aligned}$$

*The circularly polarized representation of the wave equation for the pump*

Finally, we by expressing all fields in the wave equation (30) for the temporal envelope of the pump at angular frequency  $\omega_3 = \omega_1 + \omega_2$  in the circularly polarized basis from Eq. (33) obtain<sup>9</sup>

$$\left[ \frac{\partial^2}{\partial z^2} + k_3^2 + 2ik_3k_3' \frac{\partial}{\partial t} - k_3k_3'' \frac{\partial^2}{\partial t^2} \mp i \left( \frac{\omega_3^2 \gamma_3}{c^2} + 2ia_3' \frac{\partial}{\partial t} - b_3'' \frac{\partial^2}{\partial t^2} \right) \frac{\partial}{\partial z} \right] E_{\omega_3}^{\mp} = - \left( \frac{\omega_3}{c} \right)^2 \left( p_3 \mp iq_3 \frac{\partial}{\partial z} \right) (E_{\omega_1}^{\pm} E_{\omega_2}^{\pm}). \quad (38)$$

with, just to recapitulate, the short-hand notation for the coefficients as

$$\begin{aligned} n_3 &= n(\omega_3) = [1 + \chi_{xx}(-\omega_3; \omega_3)]^{1/2}, & \gamma_3 &= \gamma_{xyz}(-\omega_3; \omega_3), \\ p_3 &= 2^{1/2} \chi_{xxx}(-\omega_3; \omega_1, \omega_2), & q_3 &= 2^{1/2} \gamma_{xyz}(-\omega_3; \omega_1, \omega_2), \\ k_3 &\equiv k(\omega_3) \equiv \frac{\omega_3 n(\omega_3)}{c}, & k_3' &\equiv k'(\omega_3) \equiv \frac{d}{d\omega} \left( \frac{\omega n(\omega)}{c} \right) \Big|_{\omega_3}, & k_3'' &\equiv k''(\omega_3) \equiv \frac{d^2}{d\omega^2} \left( \frac{\omega n(\omega)}{c} \right) \Big|_{\omega_3}. \\ a_3' &\equiv a'(\omega_3) \equiv \frac{d(k(\omega)g(\omega))}{d\omega} \Big|_{\omega_3}, & b_3'' &\equiv b''(\omega_3) \equiv \left( \frac{d^2(k(\omega)g(\omega))}{d\omega^2} - 2k'(\omega)g'(\omega) \right) \Big|_{\omega_3}, \end{aligned}$$

### Separating the total fields into their forward and backward traveling components

We will now move from the description of the total idler, signal and pump fields as described by Eqs. (36)–(38) and will from now separate them into their forward and backward traveling components. Already now, we know that the forward traveling LCP/RCP waves will follow as  $\exp(i(\omega/c)(n_k \pm \gamma_k)z)$  while the backward traveling waves will follow  $\exp(-i(\omega/c)(n_k \pm \gamma_k)z)$ ;

<sup>9</sup> In the presentation of the field equation for the pump, notice that we have the altered order “ $\mp$ ” of the circular polarizations, to match the way the pump fields enter Eqs. (36) and (37), for the idler and signal, respectively. Also notice the absence of complex conjugation of the fields involved in the interaction with the pump beam at  $\omega_3 = \omega_1 + \omega_2$ .

however, we will for the sake of simplicity, and to make the derivation step-wise, we will make the first separation as

$$E_{\omega_k}^{\pm}(z, t) = E_{\omega_k}^{f\pm}(z, t) \exp(i(\omega n_k/c)z) + E_{\omega_k}^{b\mp}(z, t) \exp(-i(\omega n_k/c)z), \quad (39)$$

for  $k = 1, 2, 3$  for the idler, signal and pump, respectively. In this separation, we will in the envelopes  $E_{\omega_k}^{f\pm}(z, t)$  and  $E_{\omega_k}^{b\pm}(z, t)$  hence still have the phase dependence from the gyrotropy ( $\gamma_k$  coefficients) present, something which we will sort out in the next step of reduction of the complexity of the wave equations for the parametric process.

By inserting the separation described by Eq. (39) into the wave equation (36) for the idler at angular frequency  $\omega_1$ , we obtain

$$\begin{aligned} & \left\{ \frac{\partial E_{\omega_1}^{f\pm}}{\partial z} \pm \frac{\omega_1 \gamma_1}{2cn_1} \left( \frac{\partial E_{\omega_1}^{f\pm}}{\partial z} + ik_1 E_{\omega_1}^{f\pm} \right) + \left( (k'_1 \mp a'_1) \frac{\partial}{\partial t} + \frac{i}{2} (k''_1 \mp b''_1) \frac{\partial^2}{\partial t^2} \right) E_{\omega_1}^{f\pm} \right. \\ & \quad \left. \pm \left( i \frac{a'_1}{k_1} \frac{\partial}{\partial t} - \frac{b''_1}{2k_1} \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_1}^{f\pm}}{\partial z} \right\} \exp \left( i \frac{\omega_1 n_1}{c} z \right) \\ & + \left\{ - \frac{\partial E_{\omega_1}^{b\mp}}{\partial z} \pm \frac{\omega_1 \gamma_1}{2cn_1} \left( \frac{\partial E_{\omega_1}^{b\mp}}{\partial z} - ik_1 E_{\omega_1}^{b\mp} \right) + \left( (k'_1 \pm a'_1) \frac{\partial}{\partial t} + \frac{i}{2} (k''_1 \pm b''_1) \frac{\partial^2}{\partial t^2} \right) E_{\omega_1}^{b\mp} \right. \\ & \quad \left. \pm \left( i \frac{a'_1}{k_1} \frac{\partial}{\partial t} - \frac{b''_1}{2k_1} \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_1}^{b\mp}}{\partial z} \right\} \exp \left( -i \frac{\omega_1 n_1}{c} z \right) \quad (40) \\ & = i \frac{\omega_1}{2cn_1} \left( p_1 \pm iq_1 \frac{\partial}{\partial z} \right) \left[ E_{\omega_3}^{f\mp} E_{\omega_2}^{f\pm*} \exp \left( i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_2 n_2}{c} \right) z \right) \right. \\ & \quad + E_{\omega_3}^{f\mp} E_{\omega_2}^{b\mp*} \exp \left( i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_2 n_2}{c} \right) z \right) \\ & \quad + E_{\omega_3}^{b\pm} E_{\omega_2}^{f\pm*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_2 n_2}{c} \right) z \right) \\ & \quad \left. + E_{\omega_3}^{b\pm} E_{\omega_2}^{b\mp*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_2 n_2}{c} \right) z \right) \right], \end{aligned}$$

while by inserting Eq. (39) into the wave equation (37) for the signal at angular frequency  $\omega_2$ ,

$$\begin{aligned} & \left\{ \frac{\partial E_{\omega_2}^{f\pm}}{\partial z} \pm \frac{\omega_2 \gamma_2}{2cn_2} \left( \frac{\partial E_{\omega_2}^{f\pm}}{\partial z} + ik_2 E_{\omega_2}^{f\pm} \right) + \left( (k'_2 \mp a'_2) \frac{\partial}{\partial t} + \frac{i}{2} (k''_2 \mp b''_2) \frac{\partial^2}{\partial t^2} \right) E_{\omega_2}^{f\pm} \right. \\ & \quad \left. \pm \left( i \frac{a'_2}{k_2} \frac{\partial}{\partial t} - \frac{b''_2}{2k_2} \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_2}^{f\pm}}{\partial z} \right\} \exp \left( i \frac{\omega_2 n_2}{c} z \right) \\ & + \left\{ - \frac{\partial E_{\omega_2}^{b\mp}}{\partial z} \pm \frac{\omega_2 \gamma_2}{2cn_2} \left( \frac{\partial E_{\omega_2}^{b\mp}}{\partial z} - ik_2 E_{\omega_2}^{b\mp} \right) + \left( (k'_2 \pm a'_2) \frac{\partial}{\partial t} + \frac{i}{2} (k''_2 \pm b''_2) \frac{\partial^2}{\partial t^2} \right) E_{\omega_2}^{b\mp} \right. \\ & \quad \left. \pm \left( i \frac{a'_2}{k_2} \frac{\partial}{\partial t} - \frac{b''_2}{2k_2} \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_2}^{b\mp}}{\partial z} \right\} \exp \left( -i \frac{\omega_2 n_2}{c} z \right) \quad (41) \\ & = i \frac{\omega_2}{2cn_2} \left( p_2 \pm iq_2 \frac{\partial}{\partial z} \right) \left[ E_{\omega_3}^{f\mp} E_{\omega_1}^{f\pm*} \exp \left( i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_1 n_1}{c} \right) z \right) \right. \\ & \quad + E_{\omega_3}^{f\mp} E_{\omega_1}^{b\mp*} \exp \left( i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_1 n_1}{c} \right) z \right) \\ & \quad + E_{\omega_3}^{b\pm} E_{\omega_1}^{f\pm*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_1 n_1}{c} \right) z \right) \\ & \quad \left. + E_{\omega_3}^{b\pm} E_{\omega_1}^{b\mp*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_1 n_1}{c} \right) z \right) \right], \end{aligned}$$

and finally, by inserting Eq. (39) into the wave equation (38) for the pump at angular frequency  $\omega_3$ ,

$$\begin{aligned}
& \left\{ \frac{\partial E_{\omega_3}^{f\mp}}{\partial z} \mp \frac{\omega_3 \gamma_3}{2cn_3} \left( \frac{\partial E_{\omega_3}^{f\mp}}{\partial z} + ik_3 E_{\omega_3}^{f\mp} \right) + \left( (k'_3 \pm a'_3) \frac{\partial}{\partial t} + \frac{i}{2} (k''_3 \pm b''_3) \frac{\partial^2}{\partial t^2} \right) E_{\omega_3}^{f\mp} \right. \\
& \quad \left. \pm \left( i \frac{a'_3}{k_3} \frac{\partial}{\partial t} - \frac{b''_3}{2k_3} \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_3}^{f\mp}}{\partial z} \right\} \exp \left( i \frac{\omega_3 n_3}{c} z \right) \\
& + \left\{ - \frac{\partial E_{\omega_3}^{b\pm}}{\partial z} \mp \frac{\omega_3 \gamma_3}{2cn_3} \left( \frac{\partial E_{\omega_3}^{b\pm}}{\partial z} - ik_3 E_{\omega_3}^{b\pm} \right) + \left( (k'_3 \mp a'_3) \frac{\partial}{\partial t} + \frac{i}{2} (k''_3 \mp b''_3) \frac{\partial^2}{\partial t^2} \right) E_{\omega_3}^{b\pm} \right. \\
& \quad \left. \mp \left( i \frac{a''_3}{k_3} \frac{\partial}{\partial t} - \frac{b''_3}{2k_3} \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_3}^{b\pm}}{\partial z} \right\} \exp \left( -i \frac{\omega_3 n_3}{c} z \right) \quad (42) \\
& = i \frac{\omega_3}{2cn_3} \left( p_3 \mp iq_3 \frac{\partial}{\partial z} \right) \left[ E_{\omega_1}^{f\pm} E_{\omega_2}^{f\pm} \exp \left( i \left( \frac{\omega_1 n_1}{c} + \frac{\omega_2 n_2}{c} \right) z \right) \right. \\
& \quad + E_{\omega_1}^{f\mp} E_{\omega_2}^{b\mp} \exp \left( i \left( \frac{\omega_1 n_1}{c} - \frac{\omega_2 n_2}{c} \right) z \right) \\
& \quad + E_{\omega_1}^{b\mp} E_{\omega_2}^{f\pm} \exp \left( -i \left( \frac{\omega_1 n_1}{c} - \frac{\omega_2 n_2}{c} \right) z \right) \\
& \quad \left. + E_{\omega_1}^{b\mp} E_{\omega_2}^{b\mp} \exp \left( -i \left( \frac{\omega_1 n_1}{c} + \frac{\omega_2 n_2}{c} \right) z \right) \right],
\end{aligned}$$

where we already from the beginning dropped the second-order spatial derivative of the fields, by applying the slowly-varying envelope approximation to the forward and backward travelling components,

$$\left| \frac{\partial^2 E_{\omega_1}^{f\pm}}{\partial z^2} \right| \ll \left| 2 \left( \frac{\omega_1 n_1}{c} \right) \frac{\partial E_{\omega_1}^{f\pm}}{\partial z} \right|, \quad \left| \frac{\partial^2 E_{\omega_1}^{b\mp}}{\partial z^2} \right| \ll \left| 2 \left( \frac{\omega_1 n_1}{c} \right) \frac{\partial E_{\omega_1}^{b\mp}}{\partial z} \right|, \quad \text{etc.} \quad (43)$$

In the rather complex coupled wave equation (40), we could in principle already at this stage start to project out terms which are closest to phase matching, in order to simplify the algebra. However, we will keep the mixed form in the right-hand side for the sake of keeping generality and allow the very same equations to be used in co-propagating as well as counter-propagating optical parametric amplification.

In the former, *co-propagating* case, we have the classical setup where the idler ( $\omega_1$ ), signal ( $\omega_2$ ) and pump ( $\omega_3$ ) fields all have their interaction taking place while propagating in the same direction. This direction may be either in the forward or backward direction, as in the case with multiply-resonant OPO. In other words, in this classic case, the involved triplet of fields in Eq. (40) for the parametric process is typically  $(E_{\omega_1}^{f\pm}, E_{\omega_2}^{f\pm}, E_{\omega_3}^{f\mp})$ .

On the other hand in the latter, *counter-propagating* case, the idler ( $\omega_1$ ) and pump ( $\omega_3$ ) propagate in, say, the positive direction, while the signal ( $\omega_2$ ) propagates in the opposite, negative direction. Thus, by keeping these options open for a while, we may accept the added complexity in algebraic handling for the sake of getting both cases covered by the same analysis and reduction of terms. In other words, in this counter-propagating case, the involved triplet of fields in Eq. (40) for the parametric process is instead typically  $(E_{\omega_1}^{f\pm}, E_{\omega_2}^{b\mp}, E_{\omega_3}^{f\mp})$ .

In addition to the standard slowly-varying envelope approximation, we may also notice that the remaining phase and amplitude changes of the remaining envelopes in any practical applications change over a length scale which is radically longer than the wavelength of light; hence we may safely assume that

$$\left| \frac{\partial E_{\omega_j}^{f\pm}}{\partial z} \right| \ll \left| \left( \frac{\omega_j n_j}{c} \right) E_{\omega_j}^{f\pm} \right|, \quad \left| \frac{\partial E_{\omega_j}^{b\mp}}{\partial z} \right| \ll \left| \left( \frac{\omega_j n_j}{c} \right) E_{\omega_j}^{b\mp} \right|, \quad (44)$$

for  $j = 1, 2, 3$ . Also, we take the opportunity to drop all cross-differential dispersive terms such as

$$\left( i \frac{a'_1}{k_1} \frac{\partial}{\partial t} - \frac{b''_1}{2k_1} \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_1}^{f\pm}}{\partial z}, \quad \left( i \frac{a'_2}{k_2} \frac{\partial}{\partial t} - \frac{b''_2}{2k_2} \frac{\partial^2}{\partial t^2} \right) \frac{\partial E_{\omega_2}^{f\pm}}{\partial z}, \quad \text{etc.}$$

Thus, just to take this somewhat cumbersome algebra step-by-step, Eq. (40) is for the idler at angular frequency  $\omega_1$  reduced to

$$\begin{aligned}
& \left\{ \frac{\partial E_{\omega_1}^{f\pm}}{\partial z} \pm i \frac{\omega_1^2 \gamma_1}{2c^2} E_{\omega_1}^{f\pm} + \left( (k_1' \mp a_1') \frac{\partial}{\partial t} + \frac{i}{2} (k_1'' \mp b_1'') \frac{\partial^2}{\partial t^2} \right) E_{\omega_1}^{f\pm} \right\} \exp \left( i \frac{\omega_1 n_1}{c} z \right) \\
& + \left\{ - \frac{\partial E_{\omega_1}^{b\mp}}{\partial z} \mp i \frac{\omega_1^2 \gamma_1}{2c^2} E_{\omega_1}^{b\mp} + \left( (k_1' \pm a_1') \frac{\partial}{\partial t} + \frac{i}{2} (k_1'' \pm b_1'') \frac{\partial^2}{\partial t^2} \right) E_{\omega_1}^{b\mp} \right\} \exp \left( -i \frac{\omega_1 n_1}{c} z \right) \\
& = i \frac{\omega_1}{2cn_1} \left( p_1 \pm iq_1 \frac{\partial}{\partial z} \right) \left[ E_{\omega_3}^{f\mp} E_{\omega_2}^{f\pm*} \exp \left( i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_2 n_2}{c} \right) z \right) \right. \\
& \quad + E_{\omega_3}^{f\mp} E_{\omega_2}^{b\mp*} \exp \left( i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_2 n_2}{c} \right) z \right) \\
& \quad + E_{\omega_3}^{b\pm} E_{\omega_2}^{f\pm*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_2 n_2}{c} \right) z \right) \\
& \quad \left. + E_{\omega_3}^{b\pm} E_{\omega_2}^{b\mp*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_2 n_2}{c} \right) z \right) \right], \tag{45}
\end{aligned}$$

while Eq. (41) for the signal at angular frequency  $\omega_2$  is reduced to

$$\begin{aligned}
& \left\{ \frac{\partial E_{\omega_2}^{f\pm}}{\partial z} \pm i \frac{\omega_2^2 \gamma_2}{2c^2} E_{\omega_2}^{f\pm} + \left( (k_2' \mp a_2') \frac{\partial}{\partial t} + \frac{i}{2} (k_2'' \mp b_2'') \frac{\partial^2}{\partial t^2} \right) E_{\omega_2}^{f\pm} \right\} \exp \left( i \frac{\omega_2 n_2}{c} z \right) \\
& + \left\{ - \frac{\partial E_{\omega_2}^{b\mp}}{\partial z} \mp i \frac{\omega_2^2 \gamma_2}{2c^2} E_{\omega_2}^{b\mp} + \left( (k_2' \pm a_2') \frac{\partial}{\partial t} + \frac{i}{2} (k_2'' \pm b_2'') \frac{\partial^2}{\partial t^2} \right) E_{\omega_2}^{b\mp} \right\} \exp \left( -i \frac{\omega_2 n_2}{c} z \right) \\
& = i \frac{\omega_2}{2cn_2} \left( p_2 \pm iq_2 \frac{\partial}{\partial z} \right) \left[ E_{\omega_3}^{f\mp} E_{\omega_1}^{f\pm*} \exp \left( i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_1 n_1}{c} \right) z \right) \right. \\
& \quad + E_{\omega_3}^{f\mp} E_{\omega_1}^{b\mp*} \exp \left( i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_1 n_1}{c} \right) z \right) \\
& \quad + E_{\omega_3}^{b\pm} E_{\omega_1}^{f\pm*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_1 n_1}{c} \right) z \right) \\
& \quad \left. + E_{\omega_3}^{b\pm} E_{\omega_1}^{b\mp*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_1 n_1}{c} \right) z \right) \right], \tag{46}
\end{aligned}$$

and, finally, that Eq. (42) for the pump at angular frequency  $\omega_3$  is reduced to

$$\begin{aligned}
& \left\{ \frac{\partial E_{\omega_3}^{f\mp}}{\partial z} \mp i \frac{\omega_3^2 \gamma_3}{2c^2} E_{\omega_3}^{f\mp} + \left( (k_3' \pm a_3') \frac{\partial}{\partial t} + \frac{i}{2} (k_3'' \pm b_3'') \frac{\partial^2}{\partial t^2} \right) E_{\omega_3}^{f\mp} \right\} \exp \left( i \frac{\omega_3 n_3}{c} z \right) \\
& + \left\{ - \frac{\partial E_{\omega_3}^{b\pm}}{\partial z} \pm i \frac{\omega_3^2 \gamma_3}{2c^2} E_{\omega_3}^{b\pm} + \left( (k_3' \mp a_3') \frac{\partial}{\partial t} + \frac{i}{2} (k_3'' \mp b_3'') \frac{\partial^2}{\partial t^2} \right) E_{\omega_3}^{b\pm} \right\} \exp \left( -i \frac{\omega_3 n_3}{c} z \right) \\
& = i \frac{\omega_3}{2cn_3} \left( p_3 \mp iq_3 \frac{\partial}{\partial z} \right) \left[ E_{\omega_1}^{f\pm} E_{\omega_2}^{f\pm} \exp \left( i \left( \frac{\omega_1 n_1}{c} + \frac{\omega_2 n_2}{c} \right) z \right) \right. \\
& \quad + E_{\omega_1}^{f\pm} E_{\omega_2}^{b\mp} \exp \left( i \left( \frac{\omega_1 n_1}{c} - \frac{\omega_2 n_2}{c} \right) z \right) \\
& \quad + E_{\omega_1}^{b\mp} E_{\omega_2}^{f\pm} \exp \left( -i \left( \frac{\omega_1 n_1}{c} - \frac{\omega_2 n_2}{c} \right) z \right) \\
& \quad \left. + E_{\omega_1}^{b\mp} E_{\omega_2}^{b\mp} \exp \left( -i \left( \frac{\omega_1 n_1}{c} + \frac{\omega_2 n_2}{c} \right) z \right) \right]. \tag{47}
\end{aligned}$$

In Eqs. (45)–(47), we may find straight away, from the appearance of the terms with coefficients  $\gamma_1$ ,  $\gamma_2$  and  $\gamma_3$ , that the gyrotropic nature of the forward and backward traveling wave envelopes can be separated by the ansatz

$$E_{\omega_k}^{f\pm}(z, t) = A_{\omega_k}^{f\pm}(z, t) \exp \left( \mp i \frac{\omega_k^2 \gamma_k}{2c^2} z \right), \tag{48a}$$

$$E_{\omega_k}^{b\mp}(z, t) = A_{\omega_k}^{b\mp}(z, t) \exp \left( \mp i \frac{\omega_k^2 \gamma_k}{2c^2} z \right), \tag{48b}$$



with  $k = 1, 2, 3$  for the idler, signal and pump, respectively. This way, carrying out the second step in the reduction of the coupled set of nonlinear PDE:s, the terms containing the gyration coefficients  $\gamma_k$  will be cancelled. Notice that in dividing the field into forward and backward traveling components, the backward traveling LCP/RCP components will be connected to the *conjugated* basis vectors  $\mathbf{e}_\pm^*$ , hence the altered order of “ $\mp$ ” in “ $E_{\omega_k}^{b\mp}$ ”.

Thus, by inserting the ansatz of Eq. (48) into Eq. (45), we for the idler at angular frequency  $\omega_1$  obtain

$$\begin{aligned} & \left\{ \frac{\partial A_{\omega_1}^{f\pm}}{\partial z} + (k'_1 \mp a'_1) \frac{\partial A_{\omega_1}^{f\pm}}{\partial t} + \frac{i}{2} (k''_1 \mp b''_1) \frac{\partial^2 A_{\omega_1}^{f\pm}}{\partial t^2} \right\} \exp \left( i \left( \frac{\omega_1 n_1}{c} \mp \frac{\omega_1^2 \gamma_1}{2c^2} \right) z \right) \\ & + \left\{ - \frac{\partial A_{\omega_1}^{b\mp}}{\partial z} + (k'_1 \pm a'_1) \frac{\partial A_{\omega_1}^{b\mp}}{\partial t} + \frac{i}{2} (k''_1 \pm b''_1) \frac{\partial^2 A_{\omega_1}^{b\mp}}{\partial t^2} \right\} \exp \left( -i \left( \frac{\omega_1 n_1}{c} z \pm \frac{\omega_1^2 \gamma_1}{2c^2} \right) z \right) \\ & = i \frac{\omega_1}{2cn_1} \left( p_1 \pm iq_1 \frac{\partial}{\partial z} \right) \left[ A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \exp \left( i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_2 n_2}{c} \right) z \pm i \left( \frac{\omega_3^2 \gamma_3}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} \right) z \right) \right. \\ & \quad + A_{\omega_3}^{f\mp} A_{\omega_2}^{b\mp*} \exp \left( i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_2 n_2}{c} \right) z \pm i \left( \frac{\omega_3^2 \gamma_3}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} \right) z \right) \\ & \quad + A_{\omega_3}^{b\pm} A_{\omega_2}^{f\pm*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_2 n_2}{c} \right) z \pm i \left( \frac{\omega_3^2 \gamma_3}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} \right) z \right) \\ & \quad \left. + A_{\omega_3}^{b\pm} A_{\omega_2}^{b\mp*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_2 n_2}{c} \right) z \pm i \left( \frac{\omega_3^2 \gamma_3}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} \right) z \right) \right], \end{aligned} \quad (49)$$

while we for the signal at angular frequency  $\omega_2$  obtain

$$\begin{aligned} & \left\{ \frac{\partial A_{\omega_2}^{f\pm}}{\partial z} + (k'_2 \mp a'_2) \frac{\partial A_{\omega_2}^{f\pm}}{\partial t} + \frac{i}{2} (k''_2 \mp b''_2) \frac{\partial^2 A_{\omega_2}^{f\pm}}{\partial t^2} \right\} \exp \left( i \left( \frac{\omega_2 n_2}{c} \mp \frac{\omega_2^2 \gamma_2}{2c^2} \right) z \right) \\ & + \left\{ - \frac{\partial A_{\omega_2}^{b\mp}}{\partial z} + (k'_2 \pm a'_2) \frac{\partial A_{\omega_2}^{b\mp}}{\partial t} + \frac{i}{2} (k''_2 \pm b''_2) \frac{\partial^2 A_{\omega_2}^{b\mp}}{\partial t^2} \right\} \exp \left( -i \left( \frac{\omega_2 n_2}{c} z \pm \frac{\omega_2^2 \gamma_2}{2c^2} \right) z \right) \\ & = i \frac{\omega_2}{2cn_2} \left( p_2 \pm iq_2 \frac{\partial}{\partial z} \right) \left[ A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \exp \left( i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_1 n_1}{c} \right) z \pm i \left( \frac{\omega_3^2 \gamma_3}{2c^2} + \frac{\omega_1^2 \gamma_1}{2c^2} \right) z \right) \right. \\ & \quad + A_{\omega_3}^{f\mp} A_{\omega_1}^{b\mp*} \exp \left( i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_1 n_1}{c} \right) z \pm i \left( \frac{\omega_3^2 \gamma_3}{2c^2} + \frac{\omega_1^2 \gamma_1}{2c^2} \right) z \right) \\ & \quad + A_{\omega_3}^{b\pm} A_{\omega_1}^{f\pm*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} + \frac{\omega_1 n_1}{c} \right) z \pm i \left( \frac{\omega_3^2 \gamma_3}{2c^2} + \frac{\omega_1^2 \gamma_1}{2c^2} \right) z \right) \\ & \quad \left. + A_{\omega_3}^{b\pm} A_{\omega_1}^{b\mp*} \exp \left( -i \left( \frac{\omega_3 n_3}{c} - \frac{\omega_1 n_1}{c} \right) z \pm i \left( \frac{\omega_3^2 \gamma_3}{2c^2} + \frac{\omega_1^2 \gamma_1}{2c^2} \right) z \right) \right], \end{aligned} \quad (50)$$

and finally for the pump at angular frequency  $\omega_3$ ,

$$\begin{aligned} & \left\{ \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} + (k'_3 \pm a'_3) \frac{\partial A_{\omega_3}^{f\mp}}{\partial t} + \frac{i}{2} (k''_3 \pm b''_3) \frac{\partial^2 A_{\omega_3}^{f\mp}}{\partial t^2} \right\} \exp \left( i \left( \frac{\omega_3 n_3}{c} \pm \frac{\omega_3^2 \gamma_3}{2c^2} \right) z \right) \\ & + \left\{ - \frac{\partial A_{\omega_3}^{b\pm}}{\partial z} + (k'_3 \mp a'_3) \frac{\partial A_{\omega_3}^{b\pm}}{\partial t} + \frac{i}{2} (k''_3 \mp b''_3) \frac{\partial^2 A_{\omega_3}^{b\pm}}{\partial t^2} \right\} \exp \left( -i \left( \frac{\omega_3 n_3}{c} z \mp \frac{\omega_3^2 \gamma_3}{2c^2} \right) z \right) \\ & = i \frac{\omega_3}{2cn_3} \left( p_3 \mp iq_3 \frac{\partial}{\partial z} \right) \left[ A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \exp \left( i \left( \frac{\omega_1 n_1}{c} + \frac{\omega_2 n_2}{c} \right) z \mp i \left( \frac{\omega_1^2 \gamma_1}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} \right) z \right) \right. \\ & \quad + A_{\omega_1}^{f\pm} A_{\omega_2}^{b\mp} \exp \left( i \left( \frac{\omega_1 n_1}{c} - \frac{\omega_2 n_2}{c} \right) z \mp i \left( \frac{\omega_1^2 \gamma_1}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} \right) z \right) \\ & \quad + A_{\omega_1}^{b\mp} A_{\omega_2}^{f\pm} \exp \left( -i \left( \frac{\omega_1 n_1}{c} - \frac{\omega_2 n_2}{c} \right) z \mp i \left( \frac{\omega_1^2 \gamma_1}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} \right) z \right) \\ & \quad \left. + A_{\omega_1}^{b\mp} A_{\omega_2}^{b\mp} \exp \left( -i \left( \frac{\omega_1 n_1}{c} + \frac{\omega_2 n_2}{c} \right) z \mp i \left( \frac{\omega_1^2 \gamma_1}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} \right) z \right) \right]. \end{aligned} \quad (51)$$

In these expressions, notice how the exponents of the right-hand sides of the idler, signal and pump equations all have the same sign alteration and same coefficients for the gyrotropic contribution to the phase evolution, for the forward as well as backward traveling components. It should here be particularly emphasized that in the right-hand sides of Eqs. (49), the spatial derivatives *operate on the entire nonlinear products acting as source terms*, including the complex-valued exponential involving the phase mismatch.

We may now assume that the idler is not self-interacting, and that we may without compromising generality project out the forward and backward traveling waves by multiplying by respective complex-valued exponents in the left-hand side and average over a few spatial periods of oscillation of the light, in the order of a wavelength. This way, we for the envelopes  $A_{\omega_k}^{f\pm} = A_{\omega_k}^{f\pm}(z, t)$  and  $A_{\omega_k}^{b\mp} = A_{\omega_k}^{b\mp}(z, t)$  of the counter-propagating waves obtain the following system, starting with the *forward traveling components* of the idler ( $\omega_1$ ), signal ( $\omega_2$ ) and pump ( $\omega_3$ ) as

$$\frac{\partial A_{\omega_1}^{f\pm}}{\partial z} + (k'_1 \mp a'_1) \frac{\partial A_{\omega_1}^{f\pm}}{\partial t} + \frac{i}{2}(k''_1 \mp b''_1) \frac{\partial^2 A_{\omega_1}^{f\pm}}{\partial t^2} \quad (52a)$$

$$\begin{aligned} &= i \frac{\omega_1}{2cn_1} \left[ p_1 A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \pm iq_1 \left( \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} A_{\omega_2}^{f\pm*} + A_{\omega_3}^{f\mp} \frac{\partial A_{\omega_2}^{f\pm*}}{\partial z} + i(k_3 - k_2 \pm (\beta_3 + \beta_2)) A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \right) \right] \\ &\quad \times \exp(i(k_3 - k_2 - k_1)z \pm i(\beta_3 + \beta_2 + \beta_1)z) \\ &+ i \frac{\omega_1}{2cn_1} \left[ p_1 A_{\omega_3}^{f\mp} A_{\omega_2}^{b\mp*} \pm iq_1 \left( \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} A_{\omega_2}^{b\mp*} + A_{\omega_3}^{f\mp} \frac{\partial A_{\omega_2}^{b\mp*}}{\partial z} + i(k_3 + k_2 \pm (\beta_3 + \beta_2)) A_{\omega_3}^{f\mp} A_{\omega_2}^{b\mp*} \right) \right] \\ &\quad \times \exp(i(k_3 + k_2 - k_1)z \pm i(\beta_3 + \beta_2 + \beta_1)z) \\ &+ i \frac{\omega_1}{2cn_1} \left[ p_1 A_{\omega_3}^{b\pm} A_{\omega_2}^{f\pm*} \pm iq_1 \left( \frac{\partial A_{\omega_3}^{b\pm}}{\partial z} A_{\omega_2}^{f\pm*} + A_{\omega_3}^{b\pm} \frac{\partial A_{\omega_2}^{f\pm*}}{\partial z} + i(-k_3 - k_2 \pm (\beta_3 + \beta_2)) A_{\omega_3}^{b\pm} A_{\omega_2}^{f\pm*} \right) \right] \\ &\quad \times \exp(-i(k_3 + k_2 + k_1)z \pm i(\beta_3 + \beta_2 + \beta_1)z) \\ &+ i \frac{\omega_1}{2cn_1} \left[ p_1 A_{\omega_3}^{b\pm} A_{\omega_2}^{b\mp*} \pm iq_1 \left( \frac{\partial A_{\omega_3}^{b\pm}}{\partial z} A_{\omega_2}^{b\mp*} + A_{\omega_3}^{b\pm} \frac{\partial A_{\omega_2}^{b\mp*}}{\partial z} + i(-k_3 + k_2 \pm (\beta_3 + \beta_2)) A_{\omega_3}^{b\pm} A_{\omega_2}^{b\mp*} \right) \right] \\ &\quad \times \exp(-i(k_3 - k_2 + k_1)z \pm i(\beta_3 + \beta_2 + \beta_1)z), \end{aligned}$$

$$\frac{\partial A_{\omega_2}^{f\pm}}{\partial z} + (k'_2 \mp a'_2) \frac{\partial A_{\omega_2}^{f\pm}}{\partial t} + \frac{i}{2}(k''_2 \mp b''_2) \frac{\partial^2 A_{\omega_2}^{f\pm}}{\partial t^2} \quad (52b)$$

$$\begin{aligned} &= i \frac{\omega_2}{2cn_2} \left[ p_2 A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \pm iq_2 \left( \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} A_{\omega_1}^{f\pm*} + A_{\omega_3}^{f\mp} \frac{\partial A_{\omega_1}^{f\pm*}}{\partial z} + i(k_3 - k_1 \pm (\beta_3 + \beta_1)) A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \right) \right] \\ &\quad \times \exp(i(k_3 - k_1 - k_2)z \pm i(\beta_3 + \beta_2 + \beta_1)z) \\ &+ i \frac{\omega_2}{2cn_2} \left[ p_2 A_{\omega_3}^{f\mp} A_{\omega_1}^{b\mp*} \pm iq_2 \left( \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} A_{\omega_1}^{b\mp*} + A_{\omega_3}^{f\mp} \frac{\partial A_{\omega_1}^{b\mp*}}{\partial z} + i(k_3 + k_1 \pm (\beta_3 + \beta_1)) A_{\omega_3}^{f\mp} A_{\omega_1}^{b\mp*} \right) \right] \\ &\quad \times \exp(i(k_3 + k_1 - k_2)z \pm i(\beta_3 + \beta_2 + \beta_1)z) \\ &+ i \frac{\omega_2}{2cn_2} \left[ p_2 A_{\omega_3}^{b\pm} A_{\omega_1}^{f\pm*} \pm iq_2 \left( \frac{\partial A_{\omega_3}^{b\pm}}{\partial z} A_{\omega_1}^{f\pm*} + A_{\omega_3}^{b\pm} \frac{\partial A_{\omega_1}^{f\pm*}}{\partial z} + i(-k_3 - k_1 \pm (\beta_3 + \beta_1)) A_{\omega_3}^{b\pm} A_{\omega_1}^{f\pm*} \right) \right] \\ &\quad \times \exp(-i(k_3 + k_1 + k_2)z \pm i(\beta_3 + \beta_2 + \beta_1)z) \\ &+ i \frac{\omega_2}{2cn_2} \left[ p_2 A_{\omega_3}^{b\pm} A_{\omega_1}^{b\mp*} \pm iq_2 \left( \frac{\partial A_{\omega_3}^{b\pm}}{\partial z} A_{\omega_1}^{b\mp*} + A_{\omega_3}^{b\pm} \frac{\partial A_{\omega_1}^{b\mp*}}{\partial z} + i(-k_3 + k_1 \pm (\beta_3 + \beta_1)) A_{\omega_3}^{b\pm} A_{\omega_1}^{b\mp*} \right) \right] \\ &\quad \times \exp(-i(k_3 - k_1 + k_2)z \pm i(\beta_3 + \beta_2 + \beta_1)z), \end{aligned}$$

$$\frac{\partial A_{\omega_3}^{f\mp}}{\partial z} + (k'_3 \pm a'_3) \frac{\partial A_{\omega_3}^{f\mp}}{\partial t} + \frac{i}{2}(k''_3 \pm b''_3) \frac{\partial^2 A_{\omega_3}^{f\mp}}{\partial t^2} \quad (52c)$$

$$\begin{aligned} &= i \frac{\omega_3}{2cn_3} \left[ p_3 A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \mp iq_3 \left( \frac{\partial A_{\omega_1}^{f\pm}}{\partial z} A_{\omega_2}^{f\pm} + A_{\omega_1}^{f\pm} \frac{\partial A_{\omega_2}^{f\pm}}{\partial z} + i(k_1 + k_2 \mp (\beta_1 + \beta_2)) A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \right) \right] \\ &\quad \times \exp(i(k_1 + k_2 - k_3)z \mp i(\beta_3 + \beta_2 + \beta_1)z) \\ &+ i \frac{\omega_3}{2cn_3} \left[ p_3 A_{\omega_1}^{f\pm} A_{\omega_2}^{b\mp} \mp iq_3 \left( \frac{\partial A_{\omega_1}^{f\pm}}{\partial z} A_{\omega_2}^{b\mp} + A_{\omega_1}^{f\pm} \frac{\partial A_{\omega_2}^{b\mp}}{\partial z} + i(k_1 - k_2 \mp (\beta_1 + \beta_2)) A_{\omega_1}^{f\pm} A_{\omega_2}^{b\mp} \right) \right] \\ &\quad \times \exp(i(k_1 - k_2 - k_3)z \mp i(\beta_3 + \beta_2 + \beta_1)z) \end{aligned}$$

$$\begin{aligned}
& + i \frac{\omega_3}{2cn_3} \left[ p_3 A_{\omega_1}^{b\mp} A_{\omega_2}^{f\pm} \mp iq_3 \left( \frac{\partial A_{\omega_1}^{b\mp}}{\partial z} A_{\omega_2}^{f\pm} + A_{\omega_1}^{b\mp} \frac{\partial A_{\omega_2}^{f\pm}}{\partial z} + i(-k_1 + k_2 \mp (\beta_1 + \beta_2)) A_{\omega_1}^{b\mp} A_{\omega_2}^{f\pm} \right) \right] \\
& \quad \times \exp(-i(k_1 - k_2 + k_3)z \mp i(\beta_3 + \beta_2 + \beta_1)z) \\
& + i \frac{\omega_3}{2cn_3} \left[ p_3 A_{\omega_1}^{b\mp} A_{\omega_2}^{b\mp} \mp iq_3 \left( \frac{\partial A_{\omega_1}^{b\mp}}{\partial z} A_{\omega_2}^{b\mp} + A_{\omega_1}^{b\mp} \frac{\partial A_{\omega_2}^{b\mp}}{\partial z} + i(-k_1 - k_2 \mp (\beta_1 + \beta_2)) A_{\omega_1}^{b\mp} A_{\omega_2}^{b\mp} \right) \right] \\
& \quad \times \exp(-i(k_1 + k_2 + k_3)z \mp i(\beta_3 + \beta_2 + \beta_1)z),
\end{aligned}$$

where we defined

$$k_j \equiv \frac{\omega_j n_j}{c}, \quad \beta_j \equiv \frac{\omega_j^2 \gamma_j}{2c^2}, \quad (53)$$

with, as previously,  $j = 1, 2, 3$  denoting the idler, signal and pump, respectively. Notice the way the phase matching in Eqs. (52) appear, with the phase mismatch from the electric-dipolar parts occurring with various combinations of the wave vector magnitudes  $k_j$ , while the gyrotropic, non-local contributions all appear as the sum  $\beta_1 + \beta_2 + \beta_3$ .

Also notice how the coefficients governing the group velocity ( $k'_j \pm a'_j$ ) and group velocity dispersion ( $k''_j \pm b''_j$ ) now always appear pair-wise with “ $\pm$ ” discriminating between the LCP/RCP components, just as for the differential phase velocity for LCP/RCP.

In similar, the *backward traveling components* are obtained from Eqs. (49)–(51) by projection, starting with the idler at angular frequency  $\omega_1$ ,

$$\begin{aligned}
& - \frac{\partial A_{\omega_1}^{b\mp}}{\partial z} + (k'_1 \pm a'_1) \frac{\partial A_{\omega_1}^{b\mp}}{\partial t} + \frac{i}{2} (k''_1 \pm b''_1) \frac{\partial^2 A_{\omega_1}^{b\mp}}{\partial t^2} \\
& = i \frac{\omega_1}{2cn_1} \left[ p_1 A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \pm iq_1 \left( \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} A_{\omega_2}^{f\pm*} + A_{\omega_3}^{f\mp} \frac{\partial A_{\omega_2}^{f\pm*}}{\partial z} + i(k_3 - k_2 \pm (\beta_3 + \beta_2)) A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \right) \right] \\
& \quad \times \exp(i(k_3 - k_2 + k_1)z \pm i(\beta_3 + \beta_2 + \beta_1)z)
\end{aligned} \quad (54a)$$

$$\begin{aligned}
& + i \frac{\omega_1}{2cn_1} \left[ p_1 A_{\omega_3}^{f\mp} A_{\omega_2}^{b\mp*} \pm iq_1 \left( \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} A_{\omega_2}^{b\mp*} + A_{\omega_3}^{f\mp} \frac{\partial A_{\omega_2}^{b\mp*}}{\partial z} + i(k_3 + k_2 \pm (\beta_3 + \beta_2)) A_{\omega_3}^{f\mp} A_{\omega_2}^{b\mp*} \right) \right] \\
& \quad \times \exp(i(k_3 + k_2 + k_1)z \pm i(\beta_3 + \beta_2 + \beta_1)z) \\
& + i \frac{\omega_1}{2cn_1} \left[ p_1 A_{\omega_3}^{b\pm} A_{\omega_2}^{f\pm*} \pm iq_1 \left( \frac{\partial A_{\omega_3}^{b\pm}}{\partial z} A_{\omega_2}^{f\pm*} + A_{\omega_3}^{b\pm} \frac{\partial A_{\omega_2}^{f\pm*}}{\partial z} + i(-k_3 - k_2 \pm (\beta_3 + \beta_2)) A_{\omega_3}^{b\pm} A_{\omega_2}^{f\pm*} \right) \right] \\
& \quad \times \exp(-i(k_3 + k_2 - k_1)z \pm i(\beta_3 + \beta_2 + \beta_1)z) \\
& + i \frac{\omega_1}{2cn_1} \left[ p_1 A_{\omega_3}^{b\pm} A_{\omega_2}^{b\mp*} \pm iq_1 \left( \frac{\partial A_{\omega_3}^{b\pm}}{\partial z} A_{\omega_2}^{b\mp*} + A_{\omega_3}^{b\pm} \frac{\partial A_{\omega_2}^{b\mp*}}{\partial z} + i(-k_3 + k_2 \pm (\beta_3 + \beta_2)) A_{\omega_3}^{b\pm} A_{\omega_2}^{b\mp*} \right) \right] \\
& \quad \times \exp(-i(k_3 - k_2 - k_1)z \pm i(\beta_3 + \beta_2 + \beta_1)z),
\end{aligned}$$

$$\begin{aligned}
& - \frac{\partial A_{\omega_2}^{b\mp}}{\partial z} + (k'_2 \pm a'_2) \frac{\partial A_{\omega_2}^{b\mp}}{\partial t} + \frac{i}{2} (k''_2 \pm b''_2) \frac{\partial^2 A_{\omega_2}^{b\mp}}{\partial t^2} \\
& = i \frac{\omega_2}{2cn_2} \left[ p_2 A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \pm iq_2 \left( \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} A_{\omega_1}^{f\pm*} + A_{\omega_3}^{f\mp} \frac{\partial A_{\omega_1}^{f\pm*}}{\partial z} + i(k_3 - k_1 \pm (\beta_3 + \beta_1)) A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \right) \right] \\
& \quad \times \exp(i(k_3 - k_1 + k_2)z \pm i(\beta_3 + \beta_2 + \beta_1)z)
\end{aligned} \quad (54b)$$

$$\begin{aligned}
& + i \frac{\omega_2}{2cn_2} \left[ p_2 A_{\omega_3}^{f\mp} A_{\omega_1}^{b\mp*} \pm iq_2 \left( \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} A_{\omega_1}^{b\mp*} + A_{\omega_3}^{f\mp} \frac{\partial A_{\omega_1}^{b\mp*}}{\partial z} + i(k_3 + k_1 \pm (\beta_3 + \beta_1)) A_{\omega_3}^{f\mp} A_{\omega_1}^{b\mp*} \right) \right] \\
& \quad \times \exp(i(k_3 + k_1 + k_2)z \pm i(\beta_3 + \beta_2 + \beta_1)z) \\
& + i \frac{\omega_2}{2cn_2} \left[ p_2 A_{\omega_3}^{b\pm} A_{\omega_1}^{f\pm*} \pm iq_2 \left( \frac{\partial A_{\omega_3}^{b\pm}}{\partial z} A_{\omega_1}^{f\pm*} + A_{\omega_3}^{b\pm} \frac{\partial A_{\omega_1}^{f\pm*}}{\partial z} + i(-k_3 - k_1 \pm (\beta_3 + \beta_1)) A_{\omega_3}^{b\pm} A_{\omega_1}^{f\pm*} \right) \right] \\
& \quad \times \exp(-i(k_3 + k_1 - k_2)z \pm i(\beta_3 + \beta_2 + \beta_1)z) \\
& + i \frac{\omega_2}{2cn_2} \left[ p_2 A_{\omega_3}^{b\pm} A_{\omega_1}^{b\mp*} \pm iq_2 \left( \frac{\partial A_{\omega_3}^{b\pm}}{\partial z} A_{\omega_1}^{b\mp*} + A_{\omega_3}^{b\pm} \frac{\partial A_{\omega_1}^{b\mp*}}{\partial z} + i(-k_3 + k_1 \pm (\beta_3 + \beta_1)) A_{\omega_3}^{b\pm} A_{\omega_1}^{b\mp*} \right) \right] \\
& \quad \times \exp(-i(k_3 - k_1 - k_2)z \pm i(\beta_3 + \beta_2 + \beta_1)z),
\end{aligned}$$

$$\begin{aligned}
& - \frac{\partial A_{\omega_3}^{b\pm}}{\partial z} + (k'_3 \mp a'_3) \frac{\partial A_{\omega_3}^{b\pm}}{\partial t} + \frac{i}{2} (k''_3 \mp b''_3) \frac{\partial^2 A_{\omega_3}^{b\pm}}{\partial t^2} \\
& \quad \times \exp(-i(k_3 - k_1 - k_2)z \pm i(\beta_3 + \beta_2 + \beta_1)z),
\end{aligned} \quad (54c)$$

$$\begin{aligned}
&= i \frac{\omega_3}{2cn_3} \left[ p_3 A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \mp iq_3 \left( \frac{\partial A_{\omega_1}^{f\pm}}{\partial z} A_{\omega_2}^{f\pm} + A_{\omega_1}^{f\pm} \frac{\partial A_{\omega_2}^{f\pm}}{\partial z} + i(k_1 + k_2 \mp (\beta_1 + \beta_2)) A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \right) \right] \\
&\quad \times \exp(i(k_1 + k_2 + k_3)z \mp i(\beta_3 + \beta_2 + \beta_1)z) \\
&+ i \frac{\omega_3}{2cn_3} \left[ p_3 A_{\omega_1}^{f\pm} A_{\omega_2}^{b\mp} \mp iq_3 \left( \frac{\partial A_{\omega_1}^{f\pm}}{\partial z} A_{\omega_2}^{b\mp} + A_{\omega_1}^{f\pm} \frac{\partial A_{\omega_2}^{b\mp}}{\partial z} + i(k_1 - k_2 \mp (\beta_1 + \beta_2)) A_{\omega_1}^{f\pm} A_{\omega_2}^{b\mp} \right) \right] \\
&\quad \times \exp(i(k_1 - k_2 + k_3)z \mp i(\beta_3 + \beta_2 + \beta_1)z) \\
&+ i \frac{\omega_3}{2cn_3} \left[ p_3 A_{\omega_1}^{b\mp} A_{\omega_2}^{f\pm} \mp iq_3 \left( \frac{\partial A_{\omega_1}^{b\mp}}{\partial z} A_{\omega_2}^{f\pm} + A_{\omega_1}^{b\mp} \frac{\partial A_{\omega_2}^{f\pm}}{\partial z} + i(-k_1 + k_2 \mp (\beta_1 + \beta_2)) A_{\omega_1}^{b\mp} A_{\omega_2}^{f\pm} \right) \right] \\
&\quad \times \exp(-i(k_1 - k_2 - k_3)z \mp i(\beta_3 + \beta_2 + \beta_1)z) \\
&+ i \frac{\omega_3}{2cn_3} \left[ p_3 A_{\omega_1}^{b\mp} A_{\omega_2}^{b\mp} \mp iq_3 \left( \frac{\partial A_{\omega_1}^{b\mp}}{\partial z} A_{\omega_2}^{b\mp} + A_{\omega_1}^{b\mp} \frac{\partial A_{\omega_2}^{b\mp}}{\partial z} + i(-k_1 - k_2 \mp (\beta_1 + \beta_2)) A_{\omega_1}^{b\mp} A_{\omega_2}^{b\mp} \right) \right] \\
&\quad \times \exp(-i(k_1 + k_2 - k_3)z \mp i(\beta_3 + \beta_2 + \beta_1)z).
\end{aligned}$$

Just as previously for the forward traveling components, the phase mismatch from the electric-dipolar parts occurring with various combinations of the wave vector magnitudes  $k_j$ , while the gyrotropic, non-local contributions all appear as the sum  $\beta_1 + \beta_2 + \beta_3$ .

### Co-directional optical parametric amplification

In the classical, co-directional configuration of optical parametric amplification, the phase matching will occur for the combination  $k_3 - k_2 - k_1 \sim 0$  of electric dipolar wave vector magnitudes. In this particular case, for the moment just focusing on the forward traveling components, the system described by Eqs. (52) will after averaging over a few spatial periods of the light reduce to

$$\begin{aligned}
&\frac{\partial A_{\omega_1}^{f\pm}}{\partial z} + (k'_1 \mp a'_1) \frac{\partial A_{\omega_1}^{f\pm}}{\partial t} + \frac{i}{2}(k''_1 \mp b''_1) \frac{\partial^2 A_{\omega_1}^{f\pm}}{\partial t^2} \\
&= i \frac{\omega_1}{2cn_1} \left[ p_1 A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \pm iq_1 \left( \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} A_{\omega_2}^{f\pm*} + A_{\omega_3}^{f\mp} \frac{\partial A_{\omega_2}^{f\pm*}}{\partial z} + i(k_3 - k_2 \pm (\beta_3 + \beta_2)) A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \right) \right] \\
&\quad \times \exp(i(k_3 - k_2 - k_1)z \pm i(\beta_3 + \beta_2 + \beta_1)z),
\end{aligned} \tag{55a}$$

$$\begin{aligned}
&\frac{\partial A_{\omega_2}^{f\pm}}{\partial z} + (k'_2 \mp a'_2) \frac{\partial A_{\omega_2}^{f\pm}}{\partial t} + \frac{i}{2}(k''_2 \mp b''_2) \frac{\partial^2 A_{\omega_2}^{f\pm}}{\partial t^2} \\
&= i \frac{\omega_2}{2cn_2} \left[ p_2 A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \pm iq_2 \left( \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} A_{\omega_1}^{f\pm*} + A_{\omega_3}^{f\mp} \frac{\partial A_{\omega_1}^{f\pm*}}{\partial z} + i(k_3 - k_1 \pm (\beta_3 + \beta_1)) A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \right) \right] \\
&\quad \times \exp(i(k_3 - k_1 - k_2)z \pm i(\beta_3 + \beta_2 + \beta_1)z),
\end{aligned} \tag{55b}$$

$$\begin{aligned}
&\frac{\partial A_{\omega_3}^{f\mp}}{\partial z} + (k'_3 \pm a'_3) \frac{\partial A_{\omega_3}^{f\mp}}{\partial t} + \frac{i}{2}(k''_3 \pm b''_3) \frac{\partial^2 A_{\omega_3}^{f\mp}}{\partial t^2} \\
&= i \frac{\omega_3}{2cn_3} \left[ p_3 A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \mp iq_3 \left( \frac{\partial A_{\omega_1}^{f\pm}}{\partial z} A_{\omega_2}^{f\pm} + A_{\omega_1}^{f\pm} \frac{\partial A_{\omega_2}^{f\pm}}{\partial z} + i(k_1 + k_2 \mp (\beta_1 + \beta_2)) A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \right) \right] \\
&\quad \times \exp(i(k_1 + k_2 - k_3)z \mp i(\beta_3 + \beta_2 + \beta_1)z),
\end{aligned} \tag{55c}$$

where the phase mismatch is described by the terms

$$\Delta\alpha \equiv k_3 - k_2 - k_1 = \frac{\omega_3 n_3}{c} - \frac{\omega_2 n_2}{c} - \frac{\omega_1 n_1}{c}, \tag{56a}$$

$$\Delta\beta \equiv \beta_3 + \beta_2 + \beta_1 = \frac{\omega_1^2 \gamma_1}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} + \frac{\omega_3^2 \gamma_3}{2c^2}. \tag{56b}$$

Again, we may in Eqs. (55) safely consider the rate of change of the field envelopes in the right-hand sides to be considerably slower than the order of the involved wavelengths,

$$\left| \frac{\partial}{\partial z} \right| \ll |k_3 - k_2|, \quad \left| \frac{\partial}{\partial z} \right| \ll |k_3 - k_1|, \quad \left| \frac{\partial}{\partial z} \right| \ll |k_1 + k_2|. \tag{57}$$

Also, we notice that the gyration coefficients  $\beta_k$  all are small in comparison to the magnitude of the wave vectors, and that we hence may assume that

$$|\beta_3 + \beta_2| \ll |k_3 - k_2|, \quad |\beta_3 + \beta_1| \ll |k_3 - k_1|, \quad |\beta_1 + \beta_2| \ll |k_1 + k_2|. \quad (58)$$

Including these simplifications, we arrive at the simplified form for the forward traveling components as

$$\begin{aligned} \frac{\partial A_{\omega_1}^{f\pm}}{\partial z} + (k'_1 \mp a'_1) \frac{\partial A_{\omega_1}^{f\pm}}{\partial t} + \frac{i}{2}(k''_1 \mp b''_1) \frac{\partial^2 A_{\omega_1}^{f\pm}}{\partial t^2} \\ = i \frac{\omega_1}{2cn_1} (p_1 \mp q_1(k_3 - k_2)) A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \exp(i(\Delta\alpha \pm \Delta\beta)z), \end{aligned} \quad (59a)$$

$$\begin{aligned} \frac{\partial A_{\omega_2}^{f\pm}}{\partial z} + (k'_2 \mp a'_2) \frac{\partial A_{\omega_2}^{f\pm}}{\partial t} + \frac{i}{2}(k''_2 \mp b''_2) \frac{\partial^2 A_{\omega_2}^{f\pm}}{\partial t^2} \\ = i \frac{\omega_2}{2cn_2} (p_2 \mp q_2(k_3 - k_1)) A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \exp(i(\Delta\alpha \pm \Delta\beta)z), \end{aligned} \quad (59b)$$

$$\begin{aligned} \frac{\partial A_{\omega_3}^{f\mp}}{\partial z} + (k'_3 \pm a'_3) \frac{\partial A_{\omega_3}^{f\mp}}{\partial t} + \frac{i}{2}(k''_3 \pm b''_3) \frac{\partial^2 A_{\omega_3}^{f\mp}}{\partial t^2} \\ = i \frac{\omega_3}{2cn_3} (p_3 \pm q_3(k_1 + k_2)) A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \exp(-i(\Delta\alpha \pm \Delta\beta)z). \end{aligned} \quad (59c)$$

### Pulsed co-directional optical parametric amplification in a moving reference frame

With a starting point in Eqs. (59), we will now concentrate on the model

$$\left( \frac{\partial}{\partial z} + (k'_1 \mp a'_1) \frac{\partial}{\partial t} + \frac{i}{2}(k''_1 \mp b''_1) \frac{\partial^2}{\partial t^2} \right) A_{\omega_1}^{f\pm} = i\kappa_1^{\pm} A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \exp(i\Delta k_{\pm}z), \quad (60a)$$

$$\left( \frac{\partial}{\partial z} + (k'_2 \mp a'_2) \frac{\partial}{\partial t} + \frac{i}{2}(k''_2 \mp b''_2) \frac{\partial^2}{\partial t^2} \right) A_{\omega_2}^{f\pm} = i\kappa_2^{\pm} A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \exp(i\Delta k_{\pm}z), \quad (60b)$$

$$\left( \frac{\partial}{\partial z} + (k'_3 \pm a'_3) \frac{\partial}{\partial t} + \frac{i}{2}(k''_3 \pm b''_3) \frac{\partial^2}{\partial t^2} \right) A_{\omega_3}^{f\mp} = i\kappa_3^{\mp} A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \exp(-i\Delta k_{\pm}z), \quad (60c)$$

where we for the notation of the coupling coefficients<sup>10</sup> adopted

$$\kappa_1^{\pm} \equiv \frac{\omega_1}{2cn_1} (p_1 \mp q_1(k_3 - k_2)), \quad \kappa_2^{\pm} \equiv \frac{\omega_2}{2cn_2} (p_2 \mp q_2(k_3 - k_1)), \quad \kappa_3^{\mp} \equiv \frac{\omega_3}{2cn_3} (p_3 \pm q_3(k_1 + k_2)), \quad (61)$$

and for the phase mismatch,

$$\begin{aligned} \Delta k_{\pm} &\equiv (\Delta\alpha \pm \Delta\beta) \\ &= \frac{\omega_3 n_3}{c} - \frac{\omega_2 n_2}{c} - \frac{\omega_1 n_1}{c} \pm \left( \frac{\omega_1^2 \gamma_1}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} + \frac{\omega_3^2 \gamma_3}{2c^2} \right). \end{aligned} \quad (62)$$

We here notice that in Eqs. (60), the group velocities  $v_i$ ,  $v_s$  and  $v_p$  of the propagating pulses for the idler  $\omega_1$ , signal  $\omega_2$  and pump  $\omega_3$  are simply given as the reciprocals of the respective coefficients  $k_j^{\pm}$ , which from the definitions in Eqs. (17), (18), and (21) are explicitly obtained as

$$v_{i\pm}^{-1} = k'_1 \mp a'_1 = \left( \frac{dk(\omega)}{d\omega} \mp \frac{d(k(\omega)g(\omega))}{d\omega} \right) \Big|_{\omega_1}, \quad (63a)$$

$$v_{s\pm}^{-1} = k'_2 \mp a'_2 = \left( \frac{dk(\omega)}{d\omega} \mp \frac{d(k(\omega)g(\omega))}{d\omega} \right) \Big|_{\omega_2}, \quad (63b)$$

$$v_{p\pm}^{-1} = k'_3 \pm a'_3 = \left( \frac{dk(\omega)}{d\omega} \pm \frac{d(k(\omega)g(\omega))}{d\omega} \right) \Big|_{\omega_3}. \quad (63c)$$

<sup>10</sup> Please observe that we here use  $\kappa_j$  (“kappa”) for the coupling coefficients, and  $k_j$ ,  $k'_j$ ,  $k''_j$ , (lower-case “k”) for the wave vector magnitude and its derivatives.

In order to reduce the coupled system described by Eqs. (60), we first of all transform it into a frame moving along with the group velocity  $v_3$  of the pump and normalize by the characteristic time of duration  $\tau$  of the pulse, by substituting the spatial coordinate  $z$  and time  $t$  for the normalized and dimensionless quantities<sup>11</sup>

$$\zeta = |k_3''|z/\tau^2, \quad s = (t - k_3'z)/\tau. \quad (64)$$

In this reference frame, keeping in mind that  $k_3' = 1/v_3$  is the inverse of the group velocity of the pump,  $s$  has the role of a *retarded time* with reference to the motion of the pump pulse.

In terms of the moving reference system  $(\zeta, s)$ , the spatial and temporal derivatives are expressed as

$$\frac{\partial}{\partial z} = \underbrace{\frac{\partial \zeta}{\partial z}}_{|k_3''|/\tau^2} \frac{\partial}{\partial \zeta} + \underbrace{\frac{\partial s}{\partial z}}_{-k_3'/\tau} \frac{\partial}{\partial s} = \frac{|k_3''|}{\tau^2} \frac{\partial}{\partial \zeta} - \frac{k_3'}{\tau} \frac{\partial}{\partial s}, \quad (65a)$$

$$\frac{\partial}{\partial t} = \underbrace{\frac{\partial \zeta}{\partial t}}_0 \frac{\partial}{\partial \zeta} + \underbrace{\frac{\partial s}{\partial t}}_{1/\tau} \frac{\partial}{\partial s} = \frac{1}{\tau} \frac{\partial}{\partial s} \Rightarrow \frac{\partial^2}{\partial t^2} = \frac{1}{\tau^2} \frac{\partial^2}{\partial s^2}, \quad (65b)$$

and the operators of the system described by Eqs. (60) are hence transformed into

$$\frac{\partial}{\partial z} + (k_1' \mp a_1') \frac{\partial}{\partial t} + i \frac{(k_1'' \mp b_1'')}{2} \frac{\partial^2}{\partial t^2} = \underbrace{\frac{|k_3''|}{\tau^2} \frac{\partial}{\partial \zeta} - \frac{k_3'}{\tau} \frac{\partial}{\partial s}}_{\frac{\partial}{\partial \zeta}} + (k_1' \mp a_1') \underbrace{\frac{1}{\tau} \frac{\partial}{\partial s}}_{\frac{\partial}{\partial t}} + i \frac{(k_1'' \mp b_1'')}{2} \underbrace{\frac{1}{\tau^2} \frac{\partial^2}{\partial s^2}}_{\frac{\partial^2}{\partial t^2}}, \quad (66a)$$

$$\frac{\partial}{\partial z} + (k_2' \mp a_2') \frac{\partial}{\partial t} + i \frac{(k_2'' \mp b_2'')}{2} \frac{\partial^2}{\partial t^2} = \underbrace{\frac{|k_3''|}{\tau^2} \frac{\partial}{\partial \zeta} - \frac{k_3'}{\tau} \frac{\partial}{\partial s}}_{\frac{\partial}{\partial \zeta}} + (k_2' \mp a_2') \underbrace{\frac{1}{\tau} \frac{\partial}{\partial s}}_{\frac{\partial}{\partial t}} + i \frac{(k_2'' \mp b_2'')}{2} \underbrace{\frac{1}{\tau^2} \frac{\partial^2}{\partial s^2}}_{\frac{\partial^2}{\partial t^2}}, \quad (66b)$$

$$\frac{\partial}{\partial z} + (k_3' \pm a_3') \frac{\partial}{\partial t} + i \frac{(k_3'' \pm b_3'')}{2} \frac{\partial^2}{\partial t^2} = \underbrace{\frac{|k_3''|}{\tau^2} \frac{\partial}{\partial \zeta} - \frac{k_3'}{\tau} \frac{\partial}{\partial s}}_{\frac{\partial}{\partial \zeta}} + (k_3' \pm a_3') \underbrace{\frac{1}{\tau} \frac{\partial}{\partial s}}_{\frac{\partial}{\partial t}} + i \frac{(k_3'' \pm b_3'')}{2} \underbrace{\frac{1}{\tau^2} \frac{\partial^2}{\partial s^2}}_{\frac{\partial^2}{\partial t^2}}, \quad (66c)$$

that is to say, by collecting the normalized and dimensional temporal ( $s$ ) and spatial ( $\zeta$ ) derivatives, we reduce Eqs. (60) into their dimensionless form<sup>12</sup>

$$\frac{|k_3''|}{\tau^2} \left( \frac{\partial}{\partial \zeta} + \frac{(k_1' \mp a_1' - k_3')\tau}{|k_3''|} \frac{\partial}{\partial s} + \frac{i}{2} \frac{(k_1'' \mp b_1'')}{|k_3''|} \frac{\partial^2}{\partial s^2} \right) A_{\omega_1}^{f\pm} = i\kappa_1^\pm A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \exp \left( i \frac{\Delta k_\pm \tau^2}{|k_3''|} \zeta \right), \quad (67a)$$

<sup>11</sup> We here follow the coordinate transformation as presented in Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, 1984), ISBN 0-471-88998-9, page 518.

<sup>12</sup> The transformation into a moving reference my means of a different time might feel like an odd and arbitrary choice. In principle, we could just as well apply a transformation where we make use of a moving spatial coordinate

$$\zeta = z + v_p t.$$

In this case, the spatial derivative will transform according to

$$\frac{\partial}{\partial z} = \underbrace{\frac{\partial \zeta}{\partial z}}_1 \frac{\partial}{\partial \zeta} + \underbrace{\frac{\partial t}{\partial z}}_{-v_p^{-1}} \frac{\partial}{\partial t} = \frac{\partial}{\partial \zeta} - \frac{1}{v_p} \frac{\partial}{\partial t},$$

which would leave the left-hand side of the system exactly in the same form as in Eqs. (66); however, in this case we would have the time entering the phase matching exponent in the right-hand side, something which clearly would mess up things somewhat. Therefore, we stick to the transformation of time rather than spatial coordinate.

$$\frac{|k_3''|}{\tau^2} \left( \frac{\partial}{\partial \zeta} + \frac{(k_2' \mp a_2' - k_3')\tau}{|k_3''|} \frac{\partial}{\partial s} + \frac{i(k_2'' \mp b_2'')}{2|k_3''|} \frac{\partial^2}{\partial s^2} \right) A_{\omega_2}^{f\pm} = i\kappa_2^\pm A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \exp\left(i \frac{\Delta k_\pm \tau^2}{|k_3''|} \zeta\right), \quad (67b)$$

$$\frac{|k_3''|}{\tau^2} \left( \frac{\partial}{\partial \zeta} \pm \frac{a_3'\tau}{|k_3''|} \frac{\partial}{\partial s} + \frac{i(k_3'' \pm b_3'')}{2|k_3''|} \frac{\partial^2}{\partial s^2} \right) A_{\omega_3}^{f\mp} = i\kappa_3^\mp A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \exp\left(-i \frac{\Delta k_\pm \tau^2}{|k_3''|} \zeta\right). \quad (67c)$$

The shape of the system described by Eqs. (67) can now be used to normalize also the field envelopes (which we recall still have the physical dimension of volts per meter). Dividing both sides by the common leading coefficient  $|k_3''|/\tau^2$  of the left-hand sides, we obtain

$$\left( \frac{\partial}{\partial \zeta} + \frac{(k_1' \mp a_1' - k_3')\tau}{|k_3''|} \frac{\partial}{\partial s} + \frac{i(k_1'' \mp b_1'')}{2|k_3''|} \frac{\partial^2}{\partial s^2} \right) A_{\omega_1}^{f\pm} = i \frac{\kappa_1^\pm \tau^2}{|k_3''|} A_{\omega_3}^{f\mp} A_{\omega_2}^{f\pm*} \exp\left(i \frac{\Delta k_\pm \tau^2}{|k_3''|} \zeta\right), \quad (68a)$$

$$\left( \frac{\partial}{\partial \zeta} + \frac{(k_2' \mp a_2' - k_3')\tau}{|k_3''|} \frac{\partial}{\partial s} + \frac{i(k_2'' \mp b_2'')}{2|k_3''|} \frac{\partial^2}{\partial s^2} \right) A_{\omega_2}^{f\pm} = i \frac{\kappa_2^\pm \tau^2}{|k_3''|} A_{\omega_3}^{f\mp} A_{\omega_1}^{f\pm*} \exp\left(i \frac{\Delta k_\pm \tau^2}{|k_3''|} \zeta\right), \quad (68b)$$

$$\left( \frac{\partial}{\partial \zeta} \pm \frac{a_3'\tau}{|k_3''|} \frac{\partial}{\partial s} + \frac{i(k_3'' \pm b_3'')}{2|k_3''|} \frac{\partial^2}{\partial s^2} \right) A_{\omega_3}^{f\mp} = i \frac{\kappa_3^\mp \tau^2}{|k_3''|} A_{\omega_1}^{f\pm} A_{\omega_2}^{f\pm} \exp\left(-i \frac{\Delta k_\pm \tau^2}{|k_3''|} \zeta\right). \quad (68c)$$

Assuming positive and real-valued coupling coefficients  $\kappa_k^\pm$ , we define the normalized and dimensionless field envelopes  $a_{\omega_j}^{f\pm}$  for the idler, signal and pump as

$$A_{\omega_1}^{f\pm} \equiv \left( \frac{\kappa_1^\pm \tau^2}{|k_3''|} \right)^{1/2} a_{\omega_1}^{f\pm}, \quad A_{\omega_2}^{f\pm} \equiv \left( \frac{\kappa_2^\pm \tau^2}{|k_3''|} \right)^{1/2} a_{\omega_2}^{f\pm}, \quad A_{\omega_3}^{f\mp} \equiv \left( \frac{\kappa_3^\mp \tau^2}{|k_3''|} \right)^{1/2} a_{\omega_3}^{f\mp}, \quad (69)$$

and further define the normalized phase mismatch related to the dimensionless and normalized spatial coordinate  $\zeta$  as

$$\begin{aligned} \Delta\phi_\pm &\equiv \Delta k_\pm \tau^2 / |k_3''| \\ &= \frac{\tau^2}{|k_3''|} \left( \frac{\omega_3 n_3}{c} - \frac{\omega_2 n_2}{c} - \frac{\omega_1 n_1}{c} \pm \left( \frac{\omega_1^2 \gamma_1}{2c^2} + \frac{\omega_2^2 \gamma_2}{2c^2} + \frac{\omega_3^2 \gamma_3}{2c^2} \right) \right). \end{aligned} \quad (70)$$

With these definitions, the normalized and dimensionless form of the system of coupled wave equations (68) takes the form

$$\left( \frac{\partial}{\partial \zeta} + \frac{(k_1' \mp a_1' - k_3')\tau}{|k_3''|} \frac{\partial}{\partial s} + \frac{i(k_1'' \mp b_1'')}{2|k_3''|} \frac{\partial^2}{\partial s^2} \right) a_{\omega_1}^{f\pm} = i\kappa_\pm a_{\omega_3}^{f\mp} a_{\omega_2}^{f\pm*} \exp(i\Delta\phi_\pm \zeta), \quad (71a)$$

$$\left( \frac{\partial}{\partial \zeta} + \frac{(k_2' \mp a_2' - k_3')\tau}{|k_3''|} \frac{\partial}{\partial s} + \frac{i(k_2'' \mp b_2'')}{2|k_3''|} \frac{\partial^2}{\partial s^2} \right) a_{\omega_2}^{f\pm} = i\kappa_\pm a_{\omega_3}^{f\mp} a_{\omega_1}^{f\pm*} \exp(i\Delta\phi_\pm \zeta), \quad (71b)$$

$$\left( \frac{\partial}{\partial \zeta} \pm \frac{a_3'\tau}{|k_3''|} \frac{\partial}{\partial s} + \frac{i(k_3'' \pm b_3'')}{2|k_3''|} \frac{\partial^2}{\partial s^2} \right) a_{\omega_3}^{f\mp} = i\kappa_\pm a_{\omega_1}^{f\pm} a_{\omega_2}^{f\pm} \exp(-i\Delta\phi_\pm \zeta), \quad (71c)$$

where the common coupling coefficient is defined in terms of the coupling coefficients of Eqs. (61) as

$$\kappa_\pm \equiv \left( \frac{\kappa_1^\pm \kappa_2^\pm \kappa_3^\mp \tau^6}{|k_3''|^3} \right)^{1/2}. \quad (72)$$

### Manley–Rowe relations for the pulsed co-directional system

In Eqs. (71), we may directly identify that the right-hand side has the same form as for the case of analysis of a continuous-wave system for the three-wave mixing. This is of course a natural consequence of that we in the nonlinear interaction consider all interaction between light and matter to be instantaneous, without any delay or spectral broadening due to the nonlinear components of the medium. All such broadening of dispersion is instead handled in the left-hand side, where the linear properties of the medium such as phase velocity, group velocity and group velocity dispersion is described in a linear part of the optical model. In this respect, we see that we may employ a similar method as in the continuous-wave case when analysing the energy balance between the idler, signal and pump, commonly denoted as the *Manley-Rowe relations*.<sup>13</sup> The extraction of the energy balance between these modes goes as follows.

#### Manley–Rowe relations in the absence of group velocity dispersion

At a first model, we skip the second derivatives in normalized time, hence dropping the group velocity dispersion from the problem. For this first case, by multiplying the left- and right-hand sides of Eqs. (71) by the respective complex conjugates of the field envelopes and adding the resulting complex conjugates of the left- and right-hand expressiond, we obtain<sup>14</sup>

$$\left(\frac{\partial}{\partial \zeta} + \frac{(k'_1 \mp a'_1 - k'_3)\tau}{|k''_3|} \frac{\partial}{\partial s}\right) |a_{\omega_1}^{f\pm}|^2 = -2\kappa_{\pm} \text{Im} [a_{\omega_3}^{f\mp} a_{\omega_2}^{f\pm*} a_{\omega_1}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta)], \quad (73a)$$

$$\left(\frac{\partial}{\partial \zeta} + \frac{(k'_2 \mp a'_2 - k'_3)\tau}{|k''_3|} \frac{\partial}{\partial s}\right) |a_{\omega_2}^{f\pm}|^2 = -2\kappa_{\pm} \text{Im} [a_{\omega_3}^{f\mp} a_{\omega_2}^{f\pm*} a_{\omega_1}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta)], \quad (73b)$$

$$\begin{aligned} \left(\frac{\partial}{\partial \zeta} \pm \frac{a'_3\tau}{|k''_3|} \frac{\partial}{\partial s}\right) |a_{\omega_3}^{f\mp}|^2 &= -2\kappa_{\pm} \text{Im} [a_{\omega_1}^{f\pm} a_{\omega_2}^{f\pm} a_{\omega_3}^{f\mp*} \exp(-i\Delta\phi_{\pm}\zeta)], \\ &= 2\kappa_{\pm} \text{Im} [a_{\omega_3}^{f\mp} a_{\omega_2}^{f\pm*} a_{\omega_1}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta)]. \end{aligned} \quad (73c)$$

By direct inspection, the idler, signal and pump intensities from the system of Eqs. (73) hence obey the energy conservation rules, or equivalently Manley–Rowe relations,

$$\left(\frac{\partial}{\partial \zeta} + \frac{(k'_1 \mp a'_1 - k'_3)\tau}{|k''_3|} \frac{\partial}{\partial s}\right) |a_{\omega_1}^{f\pm}|^2 = \left(\frac{\partial}{\partial \zeta} + \frac{(k'_2 \mp a'_2 - k'_3)\tau}{|k''_3|} \frac{\partial}{\partial s}\right) |a_{\omega_2}^{f\pm}|^2 = -\left(\frac{\partial}{\partial \zeta} \pm \frac{a'_3\tau}{|k''_3|} \frac{\partial}{\partial s}\right) |a_{\omega_3}^{f\mp}|^2 \quad (74)$$

which due to the inclusion of the relative group velocities in the coefficients of  $\partial/\partial s$  are interpreted as the rate of energy transfer between the modes during propagation along the  $\zeta$ -axis, from the presence of  $\partial/\partial \zeta$  terms, but also as the change in energy transfer rate due to any mismatch in group velocity between the pulses, from the presence of coefficients to the  $\partial/\partial s$  terms.

Notice that the Manley–Rowe relations as per above include any mismatch in group velocities between the idler-relative-pump and signal-relative-pump, as well as the differential contributions to the group velocities for LCP/RCP via the  $\mp a'_j$  terms. This is overall a signature of the purely reactive form of the medium, creating a virtual flow from a field at one frequency via the medium over to the other interacting frequencies in the current three-way mixing.

<sup>13</sup> J. M. Manley and H. E. Rowe, *Some General Properties of Nonlinear Elements – Part I: General Energy Relations*, Proceedings of the IRE, July 1956, p. 904 – 913; P. N. Butcher and D. Cotter, *The Elements of Nonlinear Optics* (Cambridge University Press, 1991), ISBN 0-521-34183-3.

<sup>14</sup> Taking into account that

$$\frac{\partial}{\partial s} |f(\zeta, s)|^2 = \frac{\partial f(\zeta, s)}{\partial s} f^*(\zeta, s) + \frac{\partial f^*(\zeta, s)}{\partial s} f(\zeta, s), \quad \text{etc.}$$

and that  $\text{Re}[if] = -\text{Im}[f]$ .



*Manley–Rowe relations in the presence of group velocity dispersion*

As for the Manley–Rowe relations in the absence of group velocity dispersion, this was a pretty straightforward and easy derivation. Let us now therefore turn our attention to the more general case in which we also include group velocity dispersion, that is to say including non-zero coefficients  $k_j''$  and  $b_j''$  from Eqs. (71c). In this case, the same multiplication by complex-conjugated field envelopes followed by an identical addition of the resulting complex conjugates leads to the form

$$\left(\frac{\partial}{\partial \zeta} + \frac{(k_1' \mp a_1' - k_3')\tau}{|k_3''|} \frac{\partial}{\partial s}\right) |a_{\omega_1}^{f\pm}|^2 + \frac{i}{2} \frac{(k_1'' \mp b_1'')}{|k_3''|} \left(\frac{\partial^2 a_{\omega_1}^{f\pm}}{\partial s^2} a_{\omega_1}^{f\pm*} - a_{\omega_1}^{f\pm} \frac{\partial^2 a_{\omega_1}^{f\pm*}}{\partial s^2}\right) = -2\kappa_{\pm} \text{Im} [a_{\omega_3}^{f\mp} a_{\omega_2}^{f\pm*} a_{\omega_1}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta)], \quad (75a)$$

$$\left(\frac{\partial}{\partial \zeta} + \frac{(k_2' \mp a_2' - k_3')\tau}{|k_3''|} \frac{\partial}{\partial s}\right) |a_{\omega_2}^{f\pm}|^2 + \frac{i}{2} \frac{(k_2'' \mp b_2'')}{|k_3''|} \left(\frac{\partial^2 a_{\omega_2}^{f\pm}}{\partial s^2} a_{\omega_2}^{f\pm*} - a_{\omega_2}^{f\pm} \frac{\partial^2 a_{\omega_2}^{f\pm*}}{\partial s^2}\right) = -2\kappa_{\pm} \text{Im} [a_{\omega_3}^{f\mp} a_{\omega_2}^{f\pm*} a_{\omega_1}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta)], \quad (75b)$$

$$\left(\frac{\partial}{\partial \zeta} \pm \frac{a_3'\tau}{|k_3''|} \frac{\partial}{\partial s}\right) a_{\omega_3}^{f\mp} + \frac{i}{2} \frac{(k_3'' \pm b_3'')}{|k_3''|} \left(\frac{\partial^2 a_{\omega_3}^{f\mp}}{\partial s^2} a_{\omega_3}^{f\mp*} - a_{\omega_3}^{f\mp} \frac{\partial^2 a_{\omega_3}^{f\mp*}}{\partial s^2}\right) = 2\kappa_{\pm} \text{Im} [a_{\omega_3}^{f\mp} a_{\omega_2}^{f\pm*} a_{\omega_1}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta)], \quad (75c)$$

which clearly is not as easy to interpret as an energy balance as the system of Eqs. (73). Here, the mixed second derivatives  $\partial^2/\partial s^2$  with an intermediate minus alters the situation compared to the one in absence of group velocity dispersion. As they currently stand in Eqs. (75), these terms do not allow for a direct interpretation in terms of derivatives of the “intensities”  $|a_{\omega_j}^{f\pm}|^2$ ,  $j = 1, 2, 3$ . However, as we via the dispersion still have not altered any premises for the medium, which still is assumed to be purely reactive in its interaction with the light, we may still expect some kind of symmetry or energy conservation rule to apply.

[ - - - TO BE CONTINUED - - - ]

**Final normalization of the system for pulsed co-directional optical parametric amplification**

It should here be emphasized that the form of Eqs. (71) with a *common* coupling coefficient opens up for a final normalization which enables us to get rid of this coupling coefficient altogether, for an even cleaner algebraic shape. We can see how this is done by multiplying both sides of the equations in the system by the common coupling coefficient  $\kappa_{\pm}$ , followed by defining the normalized field envelopes, which now are scaled by the common coupling coefficient, as

$$\tilde{a}_{\omega_1}^{f\pm}(\zeta, s) = \kappa_{\pm} a_{\omega_1}^{f\pm}(\zeta, s), \quad \tilde{a}_{\omega_2}^{f\pm}(\zeta, s) = \kappa_{\pm} a_{\omega_2}^{f\pm}(\zeta, s), \quad \tilde{a}_{\omega_3}^{f\mp}(\zeta, s) = \kappa_{\pm} a_{\omega_3}^{f\mp}(\zeta, s), \quad (76)$$

leading to Eqs. (71) taking the final normalized and dimensionless form<sup>15</sup>

<sup>15</sup> Notice that the linear part of the normalized form for the pump exactly follows the resulting Eq. (26.20) in Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, 1984), ISBN 0-471-88998-9, page 518:

$$-i \frac{\partial a}{\partial \xi} = \frac{1}{2} \left( \frac{\partial v_g}{\partial \omega} \middle/ \left| \frac{\partial v_g}{\partial \omega} \right| \right) \frac{\partial^2 a}{\partial s^2} + |a|^2 a.$$

The only difference is that here, we are concerned with a system of equations for OPA, while in Shen, the single equation concerns propagation in an optical Kerr medium, hence the “ $|a|^2 a$ ” source term in the right-hand side. Still, the approach of normalization follows analogously.

$$\left( \frac{\partial}{\partial \zeta} + \frac{(k'_1 \mp a'_1 - k'_3)\tau}{|k''_3|} \frac{\partial}{\partial s} + \frac{i(k''_1 \mp b'_1)}{2|k''_3|} \frac{\partial^2}{\partial s^2} \right) \tilde{a}_{\omega_1}^{f\pm} = i\tilde{a}_{\omega_3}^{f\mp} \tilde{a}_{\omega_2}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta), \quad (77a)$$

$$\left( \frac{\partial}{\partial \zeta} + \frac{(k'_2 \mp a'_2 - k'_3)\tau}{|k''_3|} \frac{\partial}{\partial s} + \frac{i(k''_2 \mp b'_2)}{2|k''_3|} \frac{\partial^2}{\partial s^2} \right) \tilde{a}_{\omega_2}^{f\pm} = i\tilde{a}_{\omega_3}^{f\mp} \tilde{a}_{\omega_1}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta), \quad (77b)$$

$$\left( \frac{\partial}{\partial \zeta} \pm \frac{a'_3\tau}{|k''_3|} \frac{\partial}{\partial s} + \frac{i(k''_3 \pm b'_3)}{2|k''_3|} \frac{\partial^2}{\partial s^2} \right) \tilde{a}_{\omega_3}^{f\mp} = i\tilde{a}_{\omega_1}^{f\pm} \tilde{a}_{\omega_2}^{f\pm} \exp(-i\Delta\phi_{\pm}\zeta), \quad (77c)$$

where, to recapitulate the normalization of spatial and temporal coordinates from Eq. (64),  $\zeta$  is the normalized and dimensionless spatial coordinate following the system along with the group velocity  $v_p = 1/k'_3$  of the pump and scaled against the characteristic pulse width  $\tau/v_p$  and magnitude of the group velocity dispersion  $|k''_3|$  of the pump, while  $s$  is the corresponding normalized and dimensionless time, also scaled against the characteristic pulse duration  $\tau$  of the pump.

#### The dispersion parameters

From Eq. (63), and the definition of the coefficients  $k'_k$  and  $k''_k$ ,

$$k'_k \equiv k'(\omega_k) \equiv \frac{d}{d\omega} \left( \frac{\omega n(\omega)}{c} \right) \Big|_{\omega_k}, \quad k''_k \equiv k''(\omega_k) \equiv \frac{d^2}{d\omega^2} \left( \frac{\omega n(\omega)}{c} \right) \Big|_{\omega_k},$$

for  $k = 1, 2, 3$  for the idler, signal and pump, respectively, we from the wave equation (71) have that the coefficient  $(k'_1 - k'_3)\tau/|k''_3|$  can be interpreted as the difference in reciprocal group velocities between the idler ( $v_i = 1/k'_1$ ) and pump ( $v_p = 1/k'_3$ ) pulses, normalized against the group velocity dispersion  $|k''_3|$  of the pump,

$$\frac{(k'_1 - k'_3)\tau}{|k''_3|} = \frac{\tau}{|k''_3|} \left( \frac{1}{v_i} - \frac{1}{v_p} \right).$$

Notice that the sign of the group-velocity dispersion  $k''_k$  can be either positive or negative. In the case of positive group-velocity dispersion,  $d^2k/d\omega^2 > 0$ , a long-wavelength, or equivalently low-frequency, pulse travels faster than a short-wavelength, or high-frequency, pulse.

#### Resemblance to the nonlinear Schrödinger equation

The system (71), in particular the final equation for the pump wave, slightly rearranged as

$$i \frac{\partial \tilde{a}_{\omega_3}^{f\mp}}{\partial \zeta} = \frac{1}{2} \frac{k''_3}{|k''_3|} \frac{\partial^2 \tilde{a}_{\omega_3}^{f\mp}}{\partial s^2} - \tilde{a}_{\omega_1}^{f\pm} \tilde{a}_{\omega_2}^{f\pm} \exp(-i\Delta\phi_{\pm}\zeta),$$

clearly resembles the one-dimensional nonlinear Schrödinger equation of the classic form

$$i\hbar \frac{\partial \psi(z, t)}{\partial t} = \left[ -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial z^2} + V(z, t) \right] \psi(z, t)$$

for the wave function  $\psi(z, t)$ ; however, notice that in the system of PDEs for the OPA process in Eqs. (71), the single first-derivative in the left-hand side is for the normalized *spatial coordinate*  $\zeta$  (rather than for the time  $t$  in the Schrödinger equation), while the higher derivatives are for  $s$ , being the normalized *time* (rather than for the spatial coordinate  $z$  in the Schrödinger equation).

In this respect, we may say that we have switched order of the variables between dispersion and spatial interaction; also notice that in this analogy, swapping space and time, our present potential would correspond to a potential which only varies in *time*, while it for the OPA process, as evident from Eqs. (71), instead only has a spatial dependence in terms of the nonlinear interaction and the phase mismatch factors  $\exp(i\Delta\phi_{\pm}\zeta)$  and  $\exp(-i\Delta\phi_{\pm}\zeta)$ .

### Numerical simulation of the pulsed chiral OPA process

We will now proceed with solving the normalized system of nonlinear partial differential equations (71) by means of the *Split-Step Fourier method*,<sup>1617</sup>

#### Formulation of the system of PDEs by operator formalism

In order to solve the normalized system of nonlinear partial differential equations described by the system (71) by means of the *Split-Step Fourier method*, we rewrite the system as

$$\frac{\partial \mathbf{a}(\zeta, s)}{\partial \zeta} = (\hat{\mathbb{D}} + \hat{\mathbb{N}})\mathbf{a}(\zeta, s), \quad (78)$$

where the  $3 \times 1$  column vector  $\mathbf{a}(\zeta, s)$  is defined as

$$\mathbf{a}(\zeta, s) \equiv \begin{pmatrix} \tilde{a}_{\omega_1}^{f\pm}(\zeta, s) \\ \tilde{a}_{\omega_2}^{f\pm}(\zeta, s) \\ \tilde{a}_{\omega_3}^{f\mp}(\zeta, s) \end{pmatrix} \quad (79)$$

and where the operators  $\hat{\mathbb{D}}$  and  $\hat{\mathbb{N}}$  are defined to handle the dispersion and nonlinear interactions, respectively. Here, the arguments  $(\zeta, s)$  act as the normalized version of the regular spatial coordinate and time  $(z, s)$  via Eqs. (64), with the principal difference that the normalized “time”  $s$  is expressed in a retarded time frame, effectively following the mean group velocity of the pump pulse.

Starting with the diagonal operator  $\hat{\mathbb{D}}$  for the dispersion, this is from Eqs. (77) defined in terms of the normalized time derivatives  $\partial/\partial s$  and  $\partial^2/\partial s^2$  as

$$\hat{\mathbb{D}} \equiv - \begin{pmatrix} D'_{11} \frac{\partial}{\partial s} + D''_{11} \frac{\partial^2}{\partial s^2} & 0 & 0 \\ 0 & D'_{22} \frac{\partial}{\partial s} + D''_{22} \frac{\partial^2}{\partial s^2} & 0 \\ 0 & 0 & D'_{33} \frac{\partial}{\partial s} + D''_{33} \frac{\partial^2}{\partial s^2} \end{pmatrix}, \quad (80)$$

where the coefficients of the first and second derivatives  $\partial/\partial s$  and  $\partial^2/\partial s^2$  in the normalized “time”  $s$  along the diagonal elements are

$$D'_{11}^{\pm} = \frac{(k'_1 \mp a'_1 - k'_3)\tau}{|k''_3|}, \quad D''_{11}^{\pm} = i \frac{(k''_1 \mp b''_1)}{2|k''_3|}, \quad (81a)$$

$$D'_{22}^{\pm} = \frac{(k'_2 \mp a'_2 - k'_3)\tau}{|k''_3|}, \quad D''_{22}^{\pm} = i \frac{(k''_2 \mp b''_2)}{2|k''_3|}, \quad (81b)$$

$$D'_{33}^{\pm} = \pm \frac{a'_3\tau}{|k''_3|}, \quad D''_{33}^{\pm} = i \frac{(k''_3 \pm b''_3)}{2|k''_3|}, \quad (81c)$$

where we may recall that the simplified form of the third element, for the pump, stems from the fact that we chose the group velocity of the pump as reference in the normalized time  $s$  from Eq. (64). Notice that the linear  $\hat{\mathbb{D}}$  operator only involves the normalized *time*; in just a moment, we will make use of that the time derivatives which occur in  $\hat{\mathbb{D}}$  can be expressed as a polynomial in the angular frequency in Fourier domain. It should here be stressed that the  $\hat{\mathbb{D}}$  operator has no explicit spatial dependence, and of course (due to its linearity) no dependence of the involved fields as such.

<sup>16</sup> See, for example, Govind P. Agrawal, *Nonlinear Fiber Optics* (Academic Press, 1995), ISBN 0-12-045142-5, Section 2.4.1 - *Split-Step Fourier Method*, pages 50–54.

<sup>17</sup> Ralf Deiterding, Roland Glowinski, Hilde Oliver and Stephen Poole, *A Reliable Split-Step Fourier Method for the Propagation Equation of Ultra-Fast Pulses in Single-Mode Optical Fibers*, J. Lightwave Technology **31**, 2008–2017 (2013).

Meanwhile, the nonlinear operator  $\hat{\mathbb{N}}$  is from Eqs. (77) defined as

$$\hat{\mathbb{N}} \equiv \begin{pmatrix} 0 & 0 & i\tilde{a}_{\omega_2}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta) \\ 0 & 0 & i\tilde{a}_{\omega_1}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta) \\ i\tilde{a}_{\omega_2}^{f\pm} \exp(-i\Delta\phi_{\pm}\zeta) & 0 & 0 \end{pmatrix}, \quad (82)$$

where we for the sake of simplifying the notation omitted the explicit arguments of the normalized fields  $\tilde{a}_{\omega_k}^{f\pm} = \tilde{a}_{\omega_k}^{f\pm}(\zeta, s)$ . The operator  $\hat{\mathbb{N}}$  handles the nonlinear mixing between the fields, as well as the explicit spatial dependence on the phase mismatch via the factors  $\exp(i\Delta\phi_{\pm}\zeta)$  (for the idler and signal fields) and  $\exp(-i\Delta\phi_{\pm}\zeta)$  (for the pump field).

Notice that due to the mixed-term nature of the OPA process, the definition of  $\hat{\mathbb{N}}$  is not unique; for example, we could equally well have defined the last row as instead having  $i\tilde{a}_{\omega_1}^{f\pm} \exp(-i\Delta\phi_{\pm}\zeta)$  in the second column, operating on the signal field  $\tilde{a}_{\omega_2}^{f\pm}$ , that is to say instead using

$$\hat{\mathbb{N}} \equiv \begin{pmatrix} 0 & 0 & i\tilde{a}_{\omega_2}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta) \\ 0 & 0 & i\tilde{a}_{\omega_1}^{f\pm*} \exp(i\Delta\phi_{\pm}\zeta) \\ 0 & i\tilde{a}_{\omega_1}^{f\pm} \exp(-i\Delta\phi_{\pm}\zeta) & 0 \end{pmatrix}.$$

This alternative would when multiplied by the field vector  $\mathbf{a}$  of Eq. (79) produce *exactly the same mixing term* involving the product  $\tilde{a}_{\omega_1}^{f\pm} \tilde{a}_{\omega_2}^{f\pm}$  acting as a source in the equation for the pump. However, just for the sake of sticking to one definition, we in the present analysis choose  $\hat{\mathbb{N}}$  as given by Eq. (82).

#### A scalar analogy to the operator formulation in spatial domain

As a preable to the Split-Step Fourier Method, the form of Eqs. (78) suggests that we could start off with a pedagogic check of the scalar behaviour. Thus, assume that  $D$  and  $N$  are (possibly complex-valued) constants and that  $a(z)$  is a scalar function obeying the equation with the close-to-trivial solution

$$\frac{da(z)}{dz} = (D + N)a(z) \quad \Leftrightarrow \quad a(z) = C \exp((D + N)z), \quad (83)$$

where  $C$  is a constant of integration. In other words, having obtained a solution at any spatial coordinate  $z$ , the solution of Eq. (83) at  $z + \Delta z$  is then given as

$$\begin{aligned} a(z + \Delta z) &= C \exp((D + N)(z + \Delta z)) \\ &= \underbrace{C \exp((D + N)z)}_{a(z)} \exp((D + N)\Delta z) \\ &= \exp(D\Delta z) \exp(N\Delta z) a(z). \end{aligned} \quad (84)$$

Thus, the evolution of the solution given by Eq. (84) from  $z$  to  $z + \Delta z$  can be described by a simple multiplication by  $\exp(D\Delta z) \exp(N\Delta z)$ , and it is the purpose of the Split-Step Fourier Method to perform such small steps of simulation, though considering that  $\hat{\mathbb{D}}$  and  $\hat{\mathbb{N}}$  in Eq. (78) are matrix operators, and that  $\hat{\mathbb{N}}$  in particular also has an explicit spatial and nonlinear dependence.

#### A scalar analogy to the operator formulation in spectral domain

Let us now analyze a variant of Eq. (83) in which the envelope  $a = a(z, t)$  is dependent on the spatial coordinate  $z$  and time  $t$ , and in which the operator managing the dispersion is a polynomial of time derivatives,

$$\frac{\partial a(z, t)}{\partial z} = \left[ D \left( \frac{\partial}{\partial t} \right) + N \right] a(z, t), \quad (85)$$

where  $D$  is the polynomial

$$D(X) = \sum_{k=0}^{\infty} \beta_k X^k. \quad (86)$$

If we as a first step, just as in the previous section, assume that we may treat the right-hand side as constant for a sufficiently small step  $\Delta z$ , we could then, again in analogy to Eq. (84) in the previous section, express the solution in terms of the (perhaps somewhat naive) propagator

$$a(z + \Delta z, t) = C \exp((D + N)(z + \Delta z)) = \exp\left[\Delta z D\left(\frac{\partial}{\partial t}\right)\right] \exp(N\Delta z) a(z, t). \quad (87)$$

The question is then how we should evaluate the exponential involving the time derivatives. Clearly, from a numerical perspective we should avoid splitting the envelopes in small time steps and assume that we may go ahead straight away with just computing differences and divide by the time steps. Such an approach is extremely sensitive to noise and numerical cancellation, and will lead straight to numerical inconsistencies and instabilities.

Another approach would be to observe that the exponential of a polynomial in derivatives in time, operating on an arbitrary time-dependent function  $f(t)$ , may be handled in angular frequency domain (Fourier domain) as

$$\begin{aligned} \exp\left[\Delta z D\left(\frac{\partial}{\partial t}\right)\right] f(t) &= \mathfrak{F}^{-1} \left\{ \mathfrak{F} \left[ \exp\left[\Delta z D\left(\frac{\partial}{\partial t}\right)\right] f(t) \right] \right\} \\ &= \mathfrak{F}^{-1} \left\{ \exp[\Delta z D(-i\omega)] \mathfrak{F}[f(t)] \right\} \\ &= \mathfrak{F}^{-1} \left\{ \exp[\Delta z D(-i\omega)] f(\omega) \right\}. \end{aligned} \quad (88)$$

Thus, we may from the “half-solution” expressed by Eq. (87) conclude that we may proceed in a split-step fashion as

$$a(z + \Delta z, t) = \underbrace{\mathfrak{F}^{-1} \left\{ \exp[\Delta z D(-i\omega)] \mathfrak{F} \left[ \underbrace{\exp(N\Delta z) a(z, t) \right] \right\}}_{\text{Step 2}}, \quad (89)$$

Step 1

that is to say, in the first step applying the operator  $N$  under the assumption of this being constant (in the case of a nonlinear  $N$  implying that the nonlinear contribution is treated as piece-wise constant over the small step  $\Delta z$ ), followed by the second step in which we apply the time-dependent dispersive operator  $D$  and interpret the involved time derivatives in Fourier domain.

In fact, these two steps form the very basis of the *Split-Step Fourier Method* as will now be formalised.

#### Formulation of step-wise propagation by means of the Split-Step Fourier Method

Having formulated the system of coupled and nonlinear PDEs in terms of the operator formalism described by Eqs. (78)–(82), we will now proceed with formulating the method of solution by means of the Split-Step Fourier Method. As a starter, we will describe the original formulation of the Split-Step Fourier Method, however directly afterwards we will describe a more refined version with an only marginally more complex algorithm.

**[Step 0 – Initialization]** Before entering the first step in the Split-Step Fourier Method, we initialize the involved field envelopes contained by the vector  $\mathbf{a} = \mathbf{a}(\zeta, s)$  at  $\zeta = 0$  as vectors in  $N$  samples of discretized normalized time  $s_k$ ,  $k = 0, 1, \dots, N - 1$ . As for the notation of a system of  $M$  fields contained by the  $\mathbf{a}$  column vector, we use each row to denote the  $m$ :th field envelope,

$$\mathbf{a}(\zeta, s) = \begin{pmatrix} a_0(\zeta, s) \\ a_1(\zeta, s) \\ \vdots \\ a_{M-1}(\zeta, s) \end{pmatrix}, \quad (90)$$

that is to say,  $\mathbf{a} \in \mathbb{R}^{[M \times 1]}$ . However, when discretizing this along the normalized time axis, we from a practical point allocate the field vector at normalized and dimensionless spatial coordinate

$\zeta$  as a two dimensional array with elements  $a_{m,k}$ , with as previously the  $M$  field envelopes along the row and the discrete samples in normalized and dimensionless time  $s_k$  along the columns,

$$\begin{aligned} \mathbf{a}(\zeta) &= \begin{pmatrix} a_0(\zeta, s_0) & a_0(\zeta, s_1) & a_0(\zeta, s_2) & \cdots & a_0(\zeta, s_{N-1}) \\ a_1(\zeta, s_0) & a_1(\zeta, s_1) & a_1(\zeta, s_2) & \cdots & a_1(\zeta, s_{N-1}) \\ a_2(\zeta, s_0) & a_2(\zeta, s_1) & a_2(\zeta, s_2) & \cdots & a_2(\zeta, s_{N-1}) \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ a_{M-1}(\zeta, s_0) & a_{M-1}(\zeta, s_1) & a_{M-1}(\zeta, s_2) & \cdots & a_{M-1}(\zeta, s_{N-1}) \end{pmatrix} \\ &= \begin{pmatrix} a_{0,0} & a_{0,1} & a_{0,2} & \cdots & a_{0,N-1} \\ a_{1,0} & a_{1,1} & a_{1,2} & \cdots & a_{1,N-1} \\ a_{2,0} & a_{2,1} & a_{2,2} & \cdots & a_{2,N-1} \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ a_{M-1,0} & a_{M-1,1} & a_{M-1,2} & \cdots & a_{M-1,N-1} \end{pmatrix}, \end{aligned} \quad (91)$$

that is to say,  $\mathbf{a} \in \mathbb{R}^{[M \times N]}$ . With reference to the Fourier transform appearing in the interpretation of dispersion in the scalar example of Eq. (89), we will hence in the following apply the Fourier transform row-wise of the array  $\mathbf{a}$ , separately for each field envelope  $m = 0, 1, \dots, M-1$ .

Typically, for a pulse having its peak occurring at normalized time  $s = 0$ , we would need a set of time samples properly covering the pulse before (negative time) and after (positive time) this peak. Hence, if we assume a symmetric coverage around the normalized time  $s = 0$  of its launch, we may for an even number of samples, say  $N = 2^p$  for some positive integer  $p$  (just to ensure an efficient fast Fourier transform in what is to come), and with a uniform time step  $\Delta s$  choose<sup>18</sup>

$$s_k = \left(k - \frac{(N-1)}{2}\right) \Delta s, \quad k = 0, 1, \dots, N-1, \quad (92)$$

spanning the interval  $s_{N-1} - s_0 = (N-1)\Delta s$  from start to stop. At these discrete samples of normalized time, we initialize the normalized field envelopes as described by Eq. (91) and take this as our starting point for  $\zeta = 0$ .

For a set of  $N = 2^p$  samples of normalized and dimensionless time, we will have an associated set of  $N$  samples of the corresponding normalized angular frequency, which we for simplicity will denote by  $\omega_k$ . (Despite that we usually reserve  $\omega$  as symbol for the “real” angular frequency with physical unit of rad/s.) Used in context of a Fast Fourier Transform (FFT) over  $N$  samples  $s_k$  in normalized and dimensionless time, the associated angular frequencies are<sup>19</sup>

$$\omega_k = 2\pi k f_s / N, \quad k = 0, 1, \dots, N,$$

where  $f_s = 1/\Delta s$  is the (normalized and dimensionless) temporal sample rate of the signal. As we in the dispersive step of propagation will make use of the Fourier-transformed properties of derivatives, replacing the derivatives  $\partial/\partial s$  in normalized time by multiplication by  $-i\omega$ , we will in the initialization step pre-compute these angular frequencies, as well as all powers  $w_k^n$  needed to cover the highest time-derivative present in the dispersive operator.

**[Step 1 – nonlinear propagation]** As the first step, we ignore the dispersive operator and set it to zero,

$$\hat{\mathbb{D}} = \mathbf{0} \quad \Rightarrow \quad \frac{\partial \mathbf{a}}{\partial \zeta} = \hat{\mathbb{N}} \mathbf{a}. \quad (93)$$

In other words, sort of pretend that we are dealing with an ordinary differential equation (ODE) in one single variable  $\zeta$ , albeit being a nonlinear equation in the field envelopes. For a sufficiently

<sup>18</sup> Sanity check:  $s_0 = -(N-1)\Delta s/2$  and  $s_{N-1} = (N-1)\Delta s/2$ , symmetrically placed around the centre normalized and dimensionless time  $s = 0$  as expected.

<sup>19</sup> The ordering of these frequencies depends on the programming language of choice. An array of frequencies from a given  $\Delta s$  and number of samples  $N$  can with Python’s scientific library `scipy.fftpack` be obtained by `fftshift(fftfreq(N,d=ds))`, with  $\mathbb{N} \equiv N$  and  $\mathbb{ds} \equiv \Delta s$ .

small step  $\Delta\zeta$  in the normalized spatial coordinate  $\zeta$ , we may approximate  $\hat{\mathbb{N}}$  by being constant over the small interval. In this case, an exact solution to Eq. (93) becomes

$$\mathbf{a}(\zeta + \Delta\zeta, s) = \exp(\hat{\mathbb{N}}\Delta\zeta)\mathbf{a}(\zeta, s), \quad (94)$$

in which  $\exp(\hat{\mathbb{N}}\Delta\zeta)$  should be interpreted as the *matrix exponential*;<sup>20</sup> that is to say, *not* as the common element-wise exponential. Notice that this exponential involves the envelopes of the fields present at the current position  $\zeta$ , and will change over the course of simulation; hence the exponential accordingly will need to be re-computed for each step, and cannot in general be pre-computed.

21

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<sup>20</sup> In analogy with the classic, scalar exponential function, the *matrix exponential* of a matrix  $\mathbb{X}$  can be defined as

$$\exp(\mathbb{X}) = \sum_{n=0}^{\infty} \frac{1}{n!} \mathbb{X}^n,$$

where the first term  $\mathbb{X}^0 = \mathbb{I}$  as usually is to be interpreted as the identity matrix. See, for example, <https://mathworld.wolfram.com/MatrixExponential.html> or the excellent episode *A Number to the Power of a Matrix* by Numberphile on Youtube at <https://www.youtube.com/watch?v=CHozRTwHInE>

<sup>21</sup> Notice that we here define the temporal Fourier transform  $f(\omega)$  and its inverse  $f(t)$  by

$$f(\omega) = \mathfrak{F}[f(t)] = \frac{1}{2\pi} \int_{-\infty}^{\infty} f(\tau) \exp(i\omega\tau) d\tau, \quad (95a)$$

$$f(t) = \mathfrak{F}^{-1}[f(\omega)] = \int_{-\infty}^{\infty} f(\omega) \exp(-i\omega t) d\omega, \quad (95b)$$

conforming to the standard notation in optics, say Born and Wolf.<sup>22</sup> Notice that by using this convention, derivatives in time will appear with a *negative* “ $-i\omega$ ” factor in the Fourier domain as

$$\mathfrak{F}\left[\frac{\partial f(t)}{\partial t}\right] = -i\omega \mathfrak{F}[f(t)].$$

## Example gallery of pulsed colinear OPA

### Parameters used in the simulation

The definition of the normalized length used in the simulation is from Eq. (64),

$$\zeta = |k_3''|z/\tau^2, \quad s = (t - k_3'z)/\tau,$$

interlinking the physical propagation length  $z_{\max}$ , pulse duration  $\tau$  and group velocity dispersion parameter  $|k_3''|$  for the pump. For a choice of a physical propagation length of  $z_{\max} = 10$  mm and a pulse duration of  $\tau = 10$  ps  $= 10 \times 10^{-12}$  s for the pump, which also is used as a reference for the idler and signal, a range of  $\zeta_{\max} = 1$  for the normalized length corresponds to a group dispersion parameter of  $|k_3''| = 10.0 \times 10^{-21}$  s<sup>2</sup>/m. In this respect, the group dispersion parameter is highly unknown, but we choose this value as reference. In any case, we may just as well choose a longer pulse duration  $\tau$  in case that the chosen group velocity dispersion parameter  $|k_3''|$  is found to be too low for a real medium.

For the initial pulses, we choose these all as linearly polarized, and we further choose normalized amplitudes for the fields of Eq. (77) as  $\max(\tilde{a}_{\omega_1}^{f\pm}(\zeta, 0)) = 0$  for the idler,  $\max(\tilde{a}_{\omega_2}^{f\pm}(\zeta, 0)) = 0.5$  for the signal, and  $\max(\tilde{a}_{\omega_3}^{f\pm}(\zeta, 0)) = 2.5$  for the pump. Notice that these are amplitudes, hence the intensity measures of these go as the respective *squares* of these amplitudes. The corresponding initial pulse widths are all chosen equal as 10 ps, and the pulses are also of sech(...) -shape.

For the practical implementation of the simulation, step parameters of  $N_\zeta = 4001$ ,  $\Delta\zeta = 0.0005$  for the discretized normalized spatial step in  $\zeta$ , and  $N_s = 2^{11}$ ,  $\Delta s = 0.01$  for the dicretized normalized “time”  $s$ .

We continue with the definition of the dispersion parameters of the dispersive  $\hat{D}$  operator. Notice that the sign of the group-velocity dispersion parameters  $k_j'' = d^2k_k/d\omega^2 > 0$ , a long-wavelength, or equivalently low-frequency, pulse travels faster than a short-wavelength, or high-frequency, pulse. Notice that the group velocity dispersion parameter  $|k_3''|$  for the pump *always needs to be non-zero*, due to the way that the normalization of the system of PDE:s is constructed.

The electric-dipolar part of group velocity parameters  $k_j'$  is for the pump taken as  $k_3' = 5.0 \times 10^{-9}$  s/m, corresponding to a group velocity of 2/3 of the speed of light ( $v_{g,3} = 1/k_3' = 2.0 \times 10^8$  m/s), or equivalently with a reasonable group velocity index of  $n_g = 1.5$ . For the idler and signal pulses, we assume that these travel at a speed roughly 5 percent faster than the pump, hence with  $k_1' = k_2' = 0.95 \times k_3'$ , hence with group velocities  $v_{g,1} = v_{g,2} \approx 1.05 \times v_{g,3}$ .

For the electric-dipolar part of group velocity dispersion parameters  $k_j''$  for the idler, signal and pump, we all chose these as equal, with value  $k_1'' = k_2'' = k_3'' = d(k_3')/d\omega = 10 \times 10^{-21}$  s<sup>2</sup>/m. Again, please notice that  $k_3''$  always must be non-zero, due to the way that the normlization of the coupled OPA system is constructed.

For the chiral differential corrections/contributions  $a_j$  to the group velocity parameters  $a_j'$  for idler, signal and pump, we choose these such that  $a_1' = 0.15 \times k_1'$  s/m. For the moment, this is probably an unrealistically high value, but we will currently use this for illustration of the differential group velocity between LCP/RCP modes.

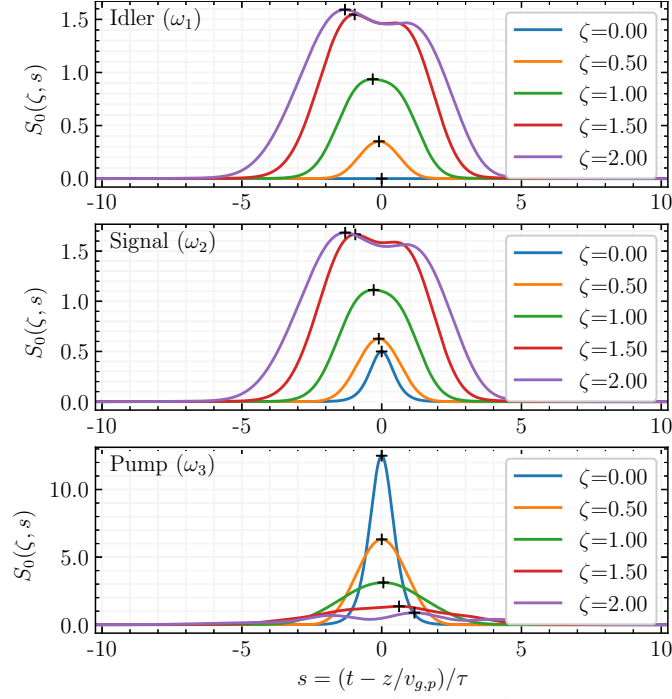
As for the chiral differential corrections/contributions to the group velocity dispersion parameters  $a_j''$  for idler, signal and pump, we choose all these as zero for the moment, as these are not the primary parameters of interest. In other words,  $a_1'' = a_2'' = a_3'' = 0$  s<sup>2</sup>/m.

Finally, for the phase mismatch, we pick values  $\delta k = 100.0$  m<sup>-1</sup> and  $\delta\alpha = 200.0$  m<sup>-1</sup>, resulting in a normalized phase mismatch from Eq. (70) of

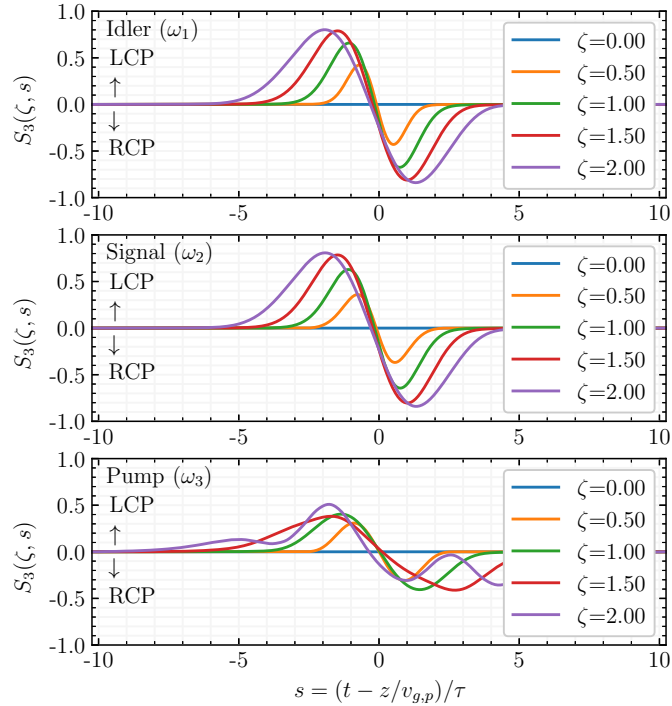
$$\Delta\phi_{\pm} \equiv (\Delta k \pm \Delta\alpha)\tau^2/|k_3''| = \frac{(10 \times 10^{-12})^2 \text{ s}^2}{10.0 \times 10^{-21} \text{ s}^2/\text{m}} \times ((100.0 \pm 200.0) \text{ m}^{-1}) = \begin{cases} 3.0 \\ -1.0 \end{cases}$$

The behaviour of this system is illustrated in the following next sections.

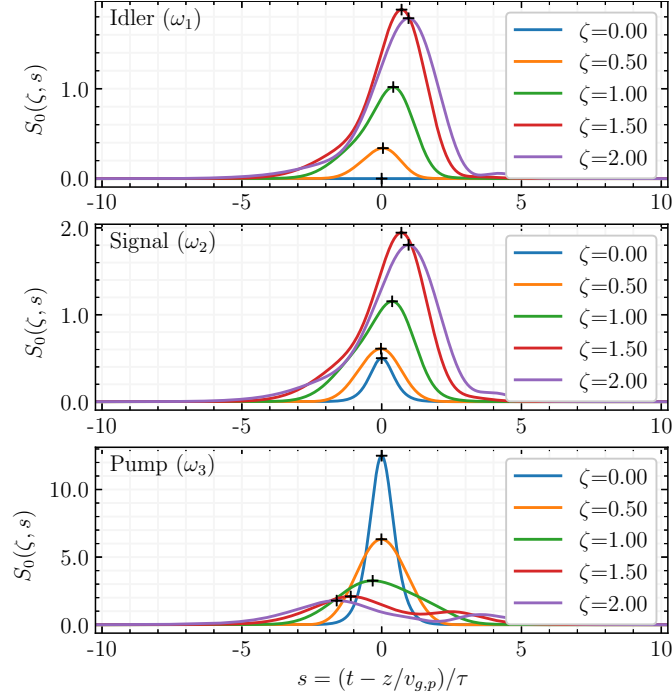


Pulse propagation with  $\Delta\alpha = 0$ 

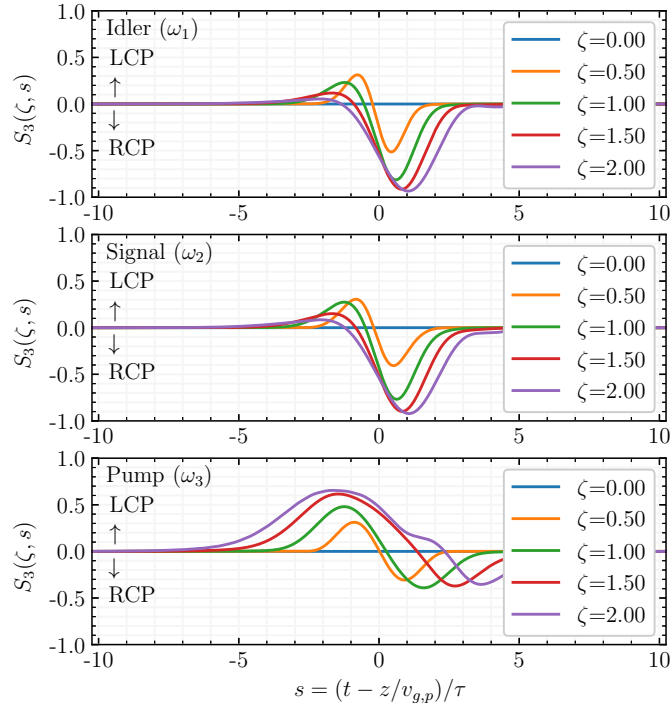
**Figure 1.** Normalized Stokes parameter  $S_0 = |a_{\omega_j}^{f+}|^2 + |a_{\omega_j}^{f-}|^2$  (total intensity) for the idler, signal and pump pulses, resolved in normalized time  $s = (t - k_3'z)/\tau$  and along normalized spatial coordinate  $\zeta = |k_3''|z/\tau^2$ , in the case where  $\Delta\alpha = 0 \text{ m}^{-1}$ .



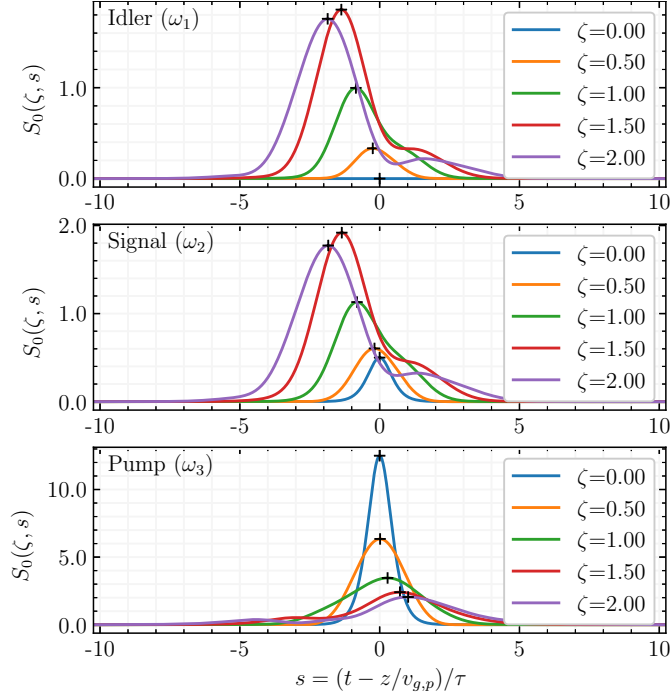
**Figure 2.** The corresponding normalized Stokes parameter  $S_3 = (|a_{\omega_j}^{f+}|^2 - |a_{\omega_j}^{f-}|^2)/S_0$  (ellipticity) for the idler, signal and pump pulses, corresponding to the same parameters as in Fig. 1.

Pulse propagation with  $\Delta\alpha > 0$ 

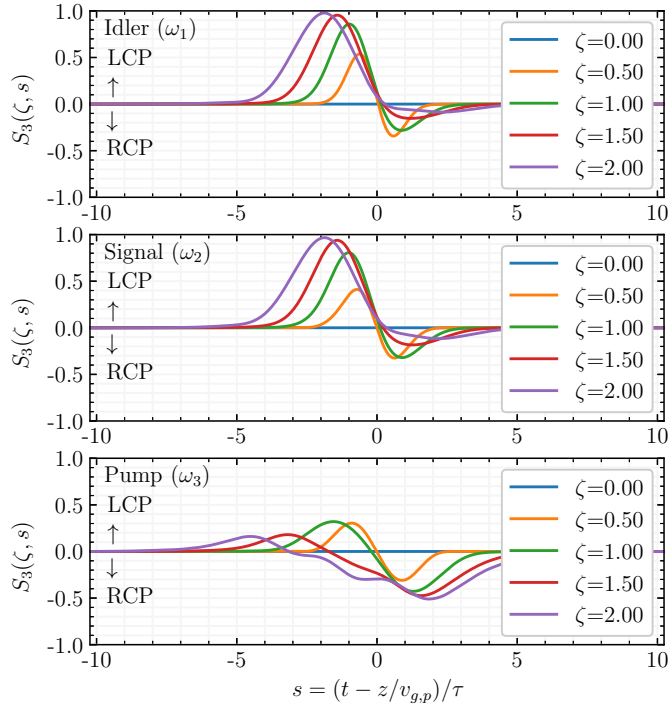
**Figure 3.** Identical parameters as for Fig. 1, with exception for a non-zero and positive  $\Delta\alpha = 200 \text{ m}^{-1}$ .



**Figure 4.** The corresponding normalized Stokes parameter  $S_3 = (|a_{\omega_j}^{f+}|^2 + |a_{\omega_j}^{f-}|^2)/S_0$  (ellipticity) for the idler, signal and pump pulses, corresponding to the same parameters as in Fig. 3.

Pulse propagation with  $\Delta\alpha > 0$ 

**Figure 5.** Identical parameters as for Figs. 1 and 3, with exception for a non-zero and negative  $\Delta\alpha = -200 \text{ m}^{-1}$ .



**Figure 6.** The corresponding normalized Stokes parameter  $S_3 = (|a_{\omega_j}^{f+}|^2 + |a_{\omega_j}^{f-}|^2)/S_0$  (ellipticity) for the idler, signal and pump pulses, corresponding to the same parameters as in Fig. 5.