Group-velocity control in the mixing of three noncollinear phase-matched waves

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A model of three-wave parametric interactions is presented, in both type I and type II noncollinear phase-matching conditions, in which the group velocities of the interacting pulses are suitably linked to each other. We consider two conditions for group velocities that are significant in the parametric generation/amplification of femtosecond pulses and determine the interaction geometry required to fulfill them in uniaxial crystals. The results are compared with those for collinear phase matching and are used to interpret the behavior of ultrabroadband parametric sources. © 1998 Optical Society of America

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1. Introduction

Solid-state sources of high-power ultrashort pulses, broadly tunable in the near-UV/visible range, are becoming more and more necessary to investigate optical nonlinearities of materials and structures newly developed for ultrafast opto-optical and opto-electronic devices. Sources based on optical parametric conversion/amplification have thus received great attention, particularly after refinements of chirped-pulse amplification techniques led to 10-fs amplifiers giving multiterawatt pulses. Harmonics of such pulses are then used either as pump pulses for parametric converters/amplifiers or in frequency-mixing schemes. They are typically mixed with the IR-tunable output of parametric generators pumped by the fundamental output of chirped-pulse amplification sources such as Ti:sapphire and Nd:glass lasers.

The large chromatic dispersion of all nonlinear crystals available for these parametric interactions, however, makes generation to UV wavelengths critical for pulse duration below 100 fs. Only accidentally do the pump wavelengths available from Ti:sapphire and Nd:glass lasers, or from their harmonics, allow phase matching (PM) with pulses at the wavelengths of interest, collinearly propagating with group velocities (GV’s) suitably matched for efficient energy exchange. For type I PM in $\beta$-BaBO$_3$ (BBO I), this occurs in spectral regions of the signal wave that, for pumping at short wavelengths, become too narrow to be useful. Broad regions in which the GV’s are mismatched by less than 100 fs/mm exist for BBO II collinearly pumped at wavelengths $\lambda_p$ between the Nd fundamental and its second harmonic. The case of the pump at the Ti:sapphire fundamental represents a particularly fortunate circumstance, recently exploited with success by Wilson and Yakovlev, in which signal and idler have opposite (and moderate) GV mismatches with respect to the pump over a reasonably broad spectral region. Wilson and Yakovlev could then amplify a femtosecond near-IR continuum with as much as 45% efficiency in 3- and 5-mm-long BBO II crystals and hence obtain broadly tunable high-energy pulses of 30–50-fs duration with frequency-conversion techniques in thin crystals.

The fact that reasonably good GV matching occurs only occasionally for collinearly propagating phase-matched pulses stimulated a number of experimental studies aimed at finding techniques of more general validity to obtain GV matching of the pump with widely tunable signal/idler pulses. It was demonstrated that, in conditions of collinear PM, the GV of a slow-traveling (extraordinary) pump can be increased, in the direction of the $k$ vector, by tilting the pulse front on the same side of the walk-off. Furthermore, a decrease in signal and idler GV’s, in the direction of the pump, so as to synchronize all pulses along the pump wave vector $k_p$, could be obtained by adopting noncollinear PM geometries.

In this paper we show that PM and GV-matching conditions, suitable for broadband interaction, can be
simultaneously verified in a number of ways. The corresponding angles for the interaction are calculated. The theoretical results are used to interpret the experimental ones mentioned above and to demonstrate the particular GV-matching conditions realized in such experiments. For example, in BBO, in which short-pulse generation was first obtained by using noncollinear PM geometries, we demonstrate that in type I PM, virtually equal GV’s in the direction of \( \mathbf{k}_p \) are obtainable over the entire tuning range \( [\lambda_p = 395 \text{ nm} \text{ (Ref}s. \text{ 6 and 7)}] \); and that in type II PM, if the walk-off is irrelevant, the GV-matching condition of actual operation \( [\lambda_p = 600 \text{ nm} \text{ (Ref} 5)] \) is GV in the direction of \( \mathbf{k}_p \), whereas GV is mismatched.

2. Phase-Matching and Group-Velocity-Matching Conditions

The basic equations of a three-wave parametric interaction occurring in the geometry depicted in Fig. 1 are the energy and momentum conservation relations:

\[
\omega_p + \omega_i = \omega_s, \tag{1}
\]

\[
k_p = k_s \cos \theta_s + k_i \cos \theta_i, \tag{2}
\]

\[
k_s \sin \theta_s = k_i \sin \theta_i, \tag{3}
\]

where

\[
k_j = \left( \omega_j / c \right) n_j, \quad j = p, s, i. \tag{4}
\]

With reference to Fig. 1, we take as positive the counterclockwise \( \theta_i \) angles, with respect to \( \mathbf{k}_p \), and the clockwise \( \theta_s \) angles, where \( \omega_s > \omega_i \). Depending on the type of PM that is realized and on the birefringence properties of the crystal, the refractive indices \( n_p, n_s, \) and \( n_i \) in Eq. (4) are related differently to the relevant dispersion relations. For uniaxial crystals one has

\[
n_p = \left[ \frac{\cos^2 \alpha + \sin^2 \alpha}{n_s^2(\omega_p)} + \frac{\sin^2 \alpha}{n_s^2(\omega_i)} \right]^{-1/2}, \tag{5}
\]

\[
n_s = n_s(\omega_s), \tag{6}
\]

\[
n_i = n_i(\omega_i) \tag{7}
\]

for type I PM \( (e \to o, o) \), whereas Eq. (7) changes into

\[
n_i = \left[ \frac{\cos^2 \alpha + \sin^2 \alpha}{n_s^2(\omega_i)} + \frac{\sin^2 \alpha}{n_s^2(\omega_i)} \right]^{-1/2} \tag{7’}
\]

for type II PM \( (e \to o, e) \).

The set of Eqs. (1)–(6) plus Eq. (7) or Eq. (7’) involves, for a given pump wavelength \( \lambda_p \), 11 unknown quantities, namely, \( \omega_s, \omega_i, k_p, k_s, k_i, \alpha, \theta_s, \theta_i, n_p, n_s, \) and \( n_i \). If we express the GV’s as

\[
GV_j = \frac{c}{n_j + \omega_j(\partial n_j/\partial \omega)_{\omega=\omega_j}}, \quad j = p, s, i, \tag{8}
\]

and add the condition of GV matching to the above system, a relation \( \alpha = \alpha(\omega) \) can be found that, in principle, provides the values of the pumping angle \( \alpha \) at which both PM and GV-matching conditions are fulfilled over the tuning (transparency) range of the crystal.

We considered two conditions of GV matching for this study: (i) that the component of GV parallel to \( \mathbf{k}_p \) be equal to GV, i.e., \( GV_{s,\text{par}} = GV_p \); and (ii) that the mismatch of GV with respect to GV be opposite that of GV, i.e., \( GV_{s,\text{par}} - GV_p = GV_p - GV_{s,\text{par}} \).

In the case of type I PM (PM I), conditions (i) and (ii) translate into the following equations, respectively:

\[
\cos \theta_i = n_i(\omega_i) + \omega_i(\partial n_i/\partial \omega)_{\omega=\omega_i}, \tag{9}
\]

\[
\cos \theta_i = \frac{1}{n_i(\omega_i) + \omega_i(\partial n_i/\partial \omega)_{\omega=\omega_i}}, \tag{10}
\]

The corresponding equations in the case of type II PM (PM II) are similar to the previous ones, except that the factor \( n_i(\omega_i) + \omega_i(\partial n_i/\partial \omega)_{\omega=\omega_i} \) appearing in Eq. (10) changes into \( n_i + \omega_i(\partial n_i/\partial \omega)_{\omega=\omega_i} \), where \( n_i \) is that given by Eq. (7’). Note that, for PM I at degeneracy, Eqs. (9) and (10) become identical and reduce to

\[
\theta_i = \cos^{-1} \left[ \frac{n_i(\omega_i/2) + \omega_i(\partial n_i/\partial \omega)_{\omega=\omega_i/2}}{n_i + \omega_i(\partial n_i/\partial \omega)_{\omega=\omega_i}} \right]. \tag{11}
\]

Therefore the corresponding solution \( \omega(\omega_i/2) \) produces \( GV_{s,\text{par}} = GV_{i,\text{par}} = GV_p \). Ideal conditions for frequency-doubling ultrashort pulses are also obtainable by using Eq. (11).

For noncollinear PM geometries, we show that the systems of Eqs. (1)–(6), (7), and (9) for PM I with \( GV_{s,\text{par}} = GV_p \) [condition (i)], Eqs. (1)–(6), (7), and (10) for PM I with \( GV_p = (GV_{s,\text{par}} + GV_{i,\text{par}})/2 \) [con-
3. **Type I Phase Matching**

The lower plots in Fig. 2 show the dependence of the pump angles \(\alpha\), satisfying Eq. (A5) [solid curves] or Eq. (A6) [dotted curves], on the signal wavelength \(\lambda_s\) for the \(\lambda_p\) values of Section 2. The upper plots in Fig. 2 show the corresponding \(\theta_s\) values. From these data information on all features that depend on the interacting-pulse GV’s can be obtained. In particular, for condition (i), that is, \(GV_{s,par} = GV_p\), we calculated the idler-to-pump GV mismatch \(GV_{ip}\) as

\[
GV_{ip} = (GV_{i,par})^{-1} - (GV_p)^{-1},
\]

and, for condition (ii), that is, \(GV_p = (GV_{s,par} + GV_{i,par})/2\), we calculated the signal-to-idler GV mismatch \(GV_{is}\) as

\[
GV_{is} = (GV_{i,par})^{-1} - (GV_{s,par})^{-1},
\]

which is the more significant, since both \(GV_{ip}\) and \(GV_{is}\) are obviously smaller. The GVM’s as calculated according to Eqs. (12) and (12’) are shown in Fig. 3 as solid and dotted curves, respectively.

The results reported in Fig. 2, which refer to PM I, allow a number of quantitative conclusions to be drawn. For BBO I the \(\alpha\) (and \(\theta_s\)) angles fulfilling the two GV-matching conditions are similar to each other over large \(\lambda_p\) intervals for all \(\lambda_p\) values, except \(\lambda_p = 790\) nm, where neither Eq. (A5) nor Eq. (A6) admits solutions. This also implies that \(GV_{i,par}\) must generally be similar to \(GV_p\) for the pumping angles that give \(GV_{s,par} = GV_p\) as confirmed by the small \(GV_{is}\) (see Fig. 3). Moreover, the \(GV_{is}\) values calculated for condition (ii), that is, \(GV_p = (GV_{s,par} + GV_{i,par})/2\), are even smaller (see Fig. 3, dotted curves). A strong coupling of the corresponding three fields is then expected if the parametric interaction occurs in a BBO I crystal at the angles \(\alpha\) and \(\theta_s\) reported in Fig. 2. Actually, the angles at which the parametric superfluorescence cone was generated and amplified under pumping with Ti:sapphire second-harmonic femtosecond pulses\(^7\) of 130-fs duration can be perfectly reproduced by using the data in Fig. 2 for \(\lambda_p = \ldots\)
395 nm. The results in Fig. 2 also agree with those, both theoretical and experimental, reported in Ref. 11 for the second and third harmonics of a Ti:sapphire fundamental pulse of 33-fs duration.

Wang et al.12 recently demonstrated generation and amplification of femtosecond pulses in the entire visible range by using 200-fs pulses at \( \lambda_p = 395 \) nm and in a collinear PM I geometry. They reported the observation of a strong off-axis parametric emission in the generation stage, which was undesired in that it caused pump depletion. Such a simultaneous presence of collinear and noncollinear generation is fully reconcilable with the results in Ref. 7: pump angles \( \alpha \) chosen on the tuning curve of collinear PM I (see the \( \lambda_s, \text{COLL} \)-versus-\( \alpha \) curves in Fig. 4) also belong to the tuning curves (almost identical) of noncollinear PM, although at another signal wavelength (\( \lambda_s, \text{NONCOLL} \) in Fig. 4). Since similar pump intensities were adopted for the generation stage in the experiments of Wang et al.12 and Di Trapani et al.,7 we can comment further on the relevance of GV mismatch, taking into account that BBO I crystals of 2- and 4-mm depths were used in Refs. 12 and 7, respectively. We calculated the time delays accumulated along the crystals by the interacting pulses and obtained the plots, also reported in Fig. 4 (right-hand scale). In the collinear case, the three delays of each pulse with respect to the others are shown in Fig. 4, although that of idler to signal (dashed curve) should play the key role; in fact, Wang et al.12 found the experimental bandwidth to agree with that, proportional to \( (\text{GVM}_{ip})^{-1} \), calculated in the approximation of negligible conversion efficiency. Their data for \( \lambda_s \) between 520 and 650 nm fit perfectly such theoretical behavior, and, although the (slight) disagreement for \( \lambda_s \) approaching degeneracy is imputable to the increase in gain that invalidates the approximation, the measured bandwidth is notably broader than expected also when \( \lambda_s < 520 \) nm. Based on the results in Fig. 4, we believe that this happens because, in this spectral region, the time delay of pump to signal is remarkably lower than that of signal to idler: when one goes toward the tuning edge, the improved synchronism of the signal with the pump pulse makes the conversion efficiency no longer negligible. The bandwidth then becomes less dependent on GVM_{ip}, which is advantageous, since the idler pulse anticipates both signal and pump pulses (dashed curve and dotted-dashed curve, re-
respectively, in Fig. 4) by time intervals that, at the crystal output, are definitely greater than the 200-fs pump duration. We observe that, in the spectral region where $GVM_{ip}$ determines the bandwidth of the collinear signal ($\lambda_p = 520-650$ nm), noncollinear superfluorescence at blue wavelengths is also phase matched. Furthermore, the noncollinear signal pulse is GV matched better to the others than is the signal collinearly generated at red wavelengths. Such a superfluorescence cone is reported in Ref. 12 as a disturbance only around the collinear signal, possibly because the collinear amplification still dominates the interaction among 150–200-fs pulses. In contrast, according to the pulse delays in Fig. 4, we expect that interacting pulses of 90–130-fs duration, such as those in Ref. 7, could be efficiently coupled only if propagating at $\theta_p$ angles to the pump equal to those reported in Fig. 2. The results of the experiment in Ref. 7 are congruous with the above discussion in that efficient noncollinear generation/amplification was obtained in that experiment by adopting $\alpha$ angles in a range out of that of collinear PM.

For the more dispersive LiIO$_3$, both GV-matching conditions can be satisfied by suitable $\alpha$ and $\theta_p$ angles for all the $\lambda_p$ values allowed by its transparency ($\lambda_p \geq 352$ nm). Figure 2 shows that the angles satisfying condition (i) are again similar to those satisfying condition (ii). As expected, the corresponding $GVM_p$ and $GVM_{ip}$ values are larger than those for BBO I, because of the greater chromatic dispersion in LiIO$_3$. Furthermore, the plots in Fig. 3 show that overall better GV-matching conditions are met by using the interaction angles satisfying Eq. (A6): 0 that is, when $GV_p = (GV_{s,par} + GV_{i,par})/2$, $GVM_{ip}$ is smaller than is $GVM_p$ when $GV_p = GV_{s,par}$. As for BBO I, we thus believe that the possibility of intense three-wave mixing in the femtosecond regime is linked to the fulfillment of condition (ii), although it is impossible to discriminate between the two GV-matching conditions (i) and (ii) by choosing the pump angle. Note that this is irrelevant in practice, because even the worst GV reported in Fig. 3 for LiIO$_3$ I is by far smaller than that of collinearly propagating pulses if the pump wavelength is in the near-UV/visible region; the GV mismatch among the interacting pulses in collinear PM is so detrimental to parametric gain, being between 1 and 2 ps/mm, that only noncollinear parametric superfluorescence was observed in a 2-cm-thick crystal under pumping at 527 nm with pulses as long as 1.1 ps. We add that our results in Fig. 2 fully agree with those in Fig. 5 of Ref. 6.

The UV-transparent material KDP is endowed with the solutions shown in Fig. 2, which extend over sizable parts of the tuning range allowed by the material transparency; for $\lambda_p \geq 527$ nm, both Eqs. (A5) and (A6) admit solutions that are appreciably different from each other, although obviously coincident at degeneracy. They give rise to the $GVM_p$ and $GVM_{ip}$ values [see Eqs. (12) and (12')] shown in Fig. 3, of which $GVM_p$ is slightly smaller. Before discussing these results, we note that $GVM_p$ is a measure of the temporal walk-off of the only GV-mismatched pulse, which is the idler, when condition (i) is fulfilled; similarly, $GVM_{ip}$ is the GV mismatch between the least synchronized pulses, i.e., signal and idler, when condition (ii) is fulfilled. In addition, since KDP is usually operated in intensity regimes in which the dynamics of the parametric interaction depends on the spatiotemporal matching of all three pulses, we calculated the three corresponding GV values for phase-matched collinearly propagating pulses ($GVM_{ip, COLL}$, $GVM_{ip, COLL}$, $GVM_{ip, COLL}$) and plotted the biggest one in magnitude, $GVM_{ip, COLL}$, in Fig. 3:

Fig. 5. Same as Fig. 2, but for PM II and either $GV_{s,par} = GV_p$ [condition (i), solid and dashed curves] or $(GV_{s,par} + GV_{i,par})/2 = GV_p$ [condition (ii), dotted curves]. The pump angle $\alpha_{COLL}$ for collinearly phase-matched waves is also shown (lower plots) for some $\lambda_p$ values.
GVM<sub>p, COLL</sub> is in all cases greater than the noncollinear GVM<sub>p</sub> and GVM<sub>ip</sub>. It is noticeable that, when our two GV-matching conditions are fulfilled at degeneracy, perfect GV matching of all pulses is achieved, which is not true for collinear PM, where the advance of the pulses at 2\(\lambda_p\), with respect to the pump is noticeable (\(\approx 130\) fs/mm). We thus believe that second-harmonic Nd:glass pulses, for example, could be more efficiently generated by sending the two fundamental pulses at approximately 2 \(\times\) 9.4° (internal angle) to each other, since \(\theta_p \equiv 9.4°\) fulfills condition (ii) at degeneracy for \(\lambda_p = 527\) nm, and by rotating the KDP I crystal so as to obtain the correct \(\alpha\) angle (see Fig. 2). It is unfortunate that this reasoning does not apply to fourth-harmonic generation, since such a perfect GV matching cannot be achieved because of the lack of solutions at degeneracy for \(\lambda_p = 263\) nm.

4. Type II Phase Matching

The plots in Fig. 5 show the dependence on \(\lambda_p\) of the pump angles \(\alpha\) and of the corresponding \(\theta_p\) values (lower and upper plots, respectively) that satisfy condition (i) (solid curves) and condition (ii) (dotted curves). The corresponding GVM’s, calculated according to Eqs. (12) and (12'), are shown in Fig. 6.

For BBO II condition (i) has one solution at \(\lambda_p = 527\) nm and two solutions (solid and dashed curves) at \(\lambda_p = 600\) nm, which give signal pulses almost collinear with the pump (\(\theta_p < 2°\)). In fact, the \(\alpha\) values in the lower plots of Fig. 5 are similar to those for collinear PM [see \(\alpha_{COLL}\) (dotted–dashed curves in Fig. 5)]. In contrast, only single solutions are found for condition (ii) at the \(\lambda_p\) values reported in Fig. 5, which are not interesting because of the large GVM<sub>ip</sub> values (see Fig. 6). We also note that the choice of a noncollinear PM geometry to synchronize the signal to the pump pulse perfectly is not really worthwhile because of the negligible GVM<sub>p, COLL</sub> differences. Furthermore, at longer pump wavelengths, where we do not find any solution, collinear PM produces GV-mismatch values below 100 fs/mm. A particularly fortunate case is that of pumping at the Ti:sapphire fundamental wavelength, in which the signal and idler pulses are mismatched by less than 50 fs/mm, on opposite sides with respect to the pump pulse, over the entire tuning range. That there is little interest in noncollinear PM geometries in BBO II is somehow confirmed by the fact that, as far as we know, the only experiment in noncollinear parametric generation is that reported in Refs. 4 and 5: an asymmetric superfluorescence cone was observed, after pumping at \(\lambda_s = 600\) nm, with greater \(\theta_p\) values when \(\theta_p < 0\), i.e., with \(\mathbf{k}_s\) between \(\mathbf{k}_p\) and the optical axis (see Fig. 1). Such an asymmetry, which is too big to be accounted for by the nonnormal incidence of the pump beam onto the crystal, is interpretable on the basis of our results. First, we note that the solution to condition (ii), reported in Fig. 5 as dotted curves for \(\lambda_p = 600\) nm, corresponds to tuning angles that, by far, are out of the range of the experimental ones. This is expected, since this solution produces GVM<sub>ip</sub> values from \(-440\) fs/mm at \(\alpha = 59.5°\) to \(-570\) fs/mm at \(\alpha = 67.5°\), which lead to an overall signal delay along the crystal (depth: 8 mm) much greater than the duration (200 fs) of the pump pulse used in the experiment.

In contrast, the two solutions that we found for condition (i), which are characterized by different GVM<sub>p</sub> (see the solid and dashed curves in Fig. 6), originate two superfluorescence cones whose apertures \(\pm\theta_p\) are plotted in Fig. 7 as a function of \(\alpha\) (upper plot). The experimental data of Refs. 4 and 5 reasonably agree with the parts of the two solutions that are evident in this figure by vertical bars. We thus ascribe the asymmetry observed in the superfluorescence emerging from the BBO II crystal to the fact that the signal emission follows different solutions for the same GV-matching condition on opposite sides of the pump beam. This behavior is not in contrast with the fact that, when the signal is generated along the cone with greater \(\theta_p\) (dashed curves in the upper plot of Fig. 7), the idler pulse is GV matched better than when it is

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig6.png}
\caption{Same as Fig. 3, but for PM II (curve styles as in Fig. 5). For BBO II and KDP II, the idler-to-pump GV mismatch for collinearly phase-matched waves, GVM<sub>p, COLL</sub>, is also reported.}
\end{figure}
generated along the cone with smaller $|\theta_s|$ (see Fig. 7, lower plot) if we take into account the lateral walk-off of both pump and idler pulse fronts (note that, since both $\alpha < \pi/2$ and $\theta_s + \alpha < \pi/2$ in all cases, the walk-off makes them drift away from the optical axis). As a result, on each cone solution, the pump and idler superimposition with the signal beam improves on the side with $\theta_s < 0$, whereas it gets worse on the opposite side. Thus, although the real system obviously adjusts to emit along the half-cone $AB$ (see Fig. 7, upper plot) because of the small (GV$ _{ip}$), as soon as PM and GV-matching conditions are fulfilled by small, still negative, signal-to-pump angles, the other solution (BC half-cone) also becomes practicable, because it is not dramatically unfavored as to GV$ _{ip}$ (see BC and BC’ in the lower plot of Fig. 7). The preference for the CD half-cone with $\theta_s > 0$, instead of for that with opposite $\theta_s$ (identical as to GV$ _{ip}$), stems from the fact that the idler walk-off causing greater divergence from $\mathbf{k}_p$ decreases the GV mismatch between the idler and pump pulses along their Poynting vectors.

For LiIO$_3$ II condition (i) has two solutions (solid and dashed curves in Fig. 5) for $\lambda_p$ between 352 and 790 nm that correspond to signal pulses with different $\theta_s$ values and increasingly deviating from $\mathbf{k}_p$ on shortening $\lambda_p$; the more off-axis solution (dashed curves) gives GV$ _{ip}$ values that are smaller in magnitude and occasionally vanish. This solution might be of interest, if one takes into account that collinear PM produces GV-mismatch values larger, although its experimental demonstration depends on the role of the inherent pump and idler walk-off. We calculated the pump and idler walk-off angles for $\lambda_p = 527$ nm and found values ranging from slightly below to slightly above those for collinear PM across the entire tuning range. Therefore walk-off should not be detrimental to the parametric amplification in this case. As for BBO II, the single acceptable solution to condition (ii), corresponding to each of the $\lambda_p$ values reported in Fig. 5, is not of interest, because of the large GV$ _{ip}$ values (see dotted curves in Fig. 6).

For KDP II as well, the existing solutions to condition (ii), which are all shown in Fig. 5, are essentially irrelevant, in that the range of phase-matchable wavelengths is heavily reduced when condition (ii) is imposed, in spite of the degrees of freedom gained by accepting noncollinear, instead of collinear, wave vectors. Condition (i) seems to be less limiting in this respect: in particular, the double solutions that we found for condition (i), plotted as solid and dashed curves in Fig. 5 for $\lambda_p = 352$ and 395 nm, allow tunability from degeneracy to the long-wavelength absorption edge. It is evident from Fig. 6, however, that in KDP II neither of the GV-matching conditions produces solutions with GV$ _{ip}$ or GV$ _{is}$ markedly smaller than those for collinear PM.

In conclusion, with the exception of only LiIO$_3$ pumped in the visible, the GV matching among the interacting pulses does not improve if noncollinear type II PM is adopted.

5. Conclusions

The group velocities of pulses undergoing three-wave parametric interactions, in both type I and type II noncollinear PM conditions, can be suitably linked to each other: we demonstrated it for the uniaxial crystals most commonly used for parametric generation/amplification at the pump wavelengths generated by available femtosecond sources. We found that operating conditions that allow either (i) GV$_p = GV _{s,par}$ or (ii) GV$_p = (GV _{s,par} + GV _{i,par})/2$ exist for all materials at various pump wavelengths of interest. In the former case, GV$_{i,par}$ is left uncontrolled; in the latter, neither GV$_{s,par}$ nor GV$_{i,par}$ is controlled, although the idler and the signal are obviously oppositely mismatched with respect to the pump. The values of GV$ _{ip}$ and GV$ _{is}$, as calculated in cases (i) and (ii), respectively, show that noncollinear PM geometries in uniaxial crystals are of interest in the case of PM I, whereas they are advantageous for only LiIO$_3$ in the case of PM II.

Appendix A

First, we solve the system of Eqs. (1)–(6), (7), and (9) that guarantee PM I and GV$_{s,par} = GV _{p}$. By deriving Eq. (5) and using Eqs. (2)–(4) in Eq. (9), we get

$$\frac{\omega_p}{2\omega_p\omega_i} \left[ Q \cos^2 \alpha + \frac{\frac{dn_s}{d\omega_p}}{n_s^3(\omega_p)} \right] = n_s(\omega_i) + \omega_i \frac{dn_s}{d\omega_i}$$

(A1)
where

\[ Q = \frac{\frac{dn_e}{d\omega_p} - \frac{dn_e}{d\omega_p}}{n_e^2(\omega_p) - n_e^2(\omega_p)} \]  

(A2)

and the following shorthand notation has been used:

\[ \frac{dn_{s,e}}{d\omega} = \left( \frac{dn_{s,e}}{d\omega} \right)_{\omega=\omega_j} . \]  

(A3)

If we define

\[ D = \frac{1}{n_e^2(\omega_p)} - \frac{1}{n_e^2(\omega_p)} \]  

(A4)

and substitute Eqs. (5)–(7) into Eq. (A1), this becomes

\[ \left\{ D[\omega_s^2 n_s^2(\omega_s) - \omega_s^2 n_s^2(\omega_s)] \left( \frac{D}{\omega_p} + Q \right) - 2D^2 \omega_s n_s(\omega_s) \left[ n_s(\omega_s) + \omega_s \frac{dn_e}{d\omega_p} \right] \right\} \cos^4 \alpha \]

\[ + \left\{ \omega_p^2 \left( \frac{D}{\omega_p} + Q \right) + \frac{\omega_s^2 n_s^2(\omega_s) - \omega_s^2 n_s^2(\omega_s)}{n_s^2(\omega_p)} \left( \frac{2D}{\omega_p} + Q \right) \right\} \]

\[ + D[\omega_s^2 n_s^2(\omega_s) - \omega_s^2 n_s^2(\omega_s)] \frac{\frac{dn_e}{d\omega_p}}{n_s^2(\omega_p)} - \frac{4D}{n_s^2(\omega_p)} \omega_s n_s(\omega_s) \left[ n_s(\omega_s) + \omega_s \frac{dn_e}{d\omega_p} \right] \cos^4 \alpha \]

\[ + \frac{\omega_p^2}{n_e^2(\omega_p)} + \frac{\omega_p}{n_e^2(\omega_p)} + \frac{\omega_s^2 n_s^2(\omega_s) - \omega_s^2 n_s^2(\omega_s)}{n_s^2(\omega_p)} \frac{dn_e}{d\omega_p} \]

\[ -2\omega_s n_s(\omega_s) \left[ n_s(\omega_s) + \omega_s \frac{dn_e}{d\omega_p} \right] + \frac{\omega_p^2}{n_e^2(\omega_p)} = 0. \]  

(A5)

The condition in which the signal- and the idler-pulse lags with respect to the pump pulse is opposite, that is, condition (ii), is described by the set of Eqs. (1)–(7) plus Eq. (9). Through substitutions similar to those used to obtain Eq. (A5) from Eq. (9), Eq. (10) can be transformed into

\[ \left\{ D[\omega_s^2 n_s^2(\omega_s) - \omega_s^2 n_s^2(\omega_s)] M \left( \frac{D}{\omega_p} + Q \right) - 4D^2 \right\} \cos^4 \alpha \]

\[ + \left\{ \omega_p^2 N \left( \frac{D}{\omega_p} + Q \right) + \frac{\omega_s^2 n_s^2(\omega_s) - \omega_s^2 n_s^2(\omega_s)}{n_s^2(\omega_p)} M \left( \frac{2D}{\omega_p} + Q \right) \right\} \cos^4 \alpha \]

\[ + D[\omega_s^2 n_s^2(\omega_s) - \omega_s^2 n_s^2(\omega_s)] M \frac{\frac{dn_e}{d\omega_p}}{n_s^2(\omega_p)} - \frac{8D}{n_s^2(\omega_p)} \]  

where we defined

\[ M = \frac{1}{\omega_s n_s(\omega_s) + \omega_s \left( \frac{dn_e}{d\omega_p} \right)_{\omega=\omega_p} + \frac{1}{\omega_s n_s(\omega_s) + \omega_s \left( \frac{dn_e}{d\omega_p} \right)_{\omega=\omega_p}}}. \]  

(A7)

\[ N = \frac{1}{\omega_s n_s(\omega_s) + \omega_s \left( \frac{dn_e}{d\omega_p} \right)_{\omega=\omega_p} + \frac{1}{\omega_s n_s(\omega_s) + \omega_s \left( \frac{dn_e}{d\omega_p} \right)_{\omega=\omega_p}}}. \]  

(A8)
If we take into account Eq. (1) and the definitions in Eqs. (A2)–(A4), (A7), and (A8), Eqs. (A5) and (A6) are the desired ones.

**Appendix B**

We solved the system of Eqs. (1)–(6), (7 ′), and (9) that guarantee PM II and GV\_(\nu) = GV\_p. As a first step, we eliminated \( \theta_i \) from Eq. (7 ′) by using Eqs. (2)–(4) and obtained

\[
A(\omega_i^2 n_p^2 - \omega_i^2 n_s^2 + \omega_i^2 n_t^2) + C4\omega_i^2 n_i^2 \omega_p^2 n_p^2 - D\omega_i^2 = B(\omega_i^2 n_p^2 - \omega_i^2 n_s^2 + \omega_i^2 n_i^2)[4\omega_i^2 n_i^2 \omega_p^2 n_p^2 - (\omega_i^2 n_p^2 - \omega_i^2 n_s^2 + \omega_i^2 n_t^2)^{1/2}],
\]

where

\[
A = n_i^2(\omega)D_i(\cos^2 \alpha - \sin^2 \alpha),
B = 2n_i^2(\omega)D_i \sin \alpha \cos \alpha,
C = n_i^2(\omega)D_i \sin^2 \alpha + 1,
D = n_i^2(\omega),
\]

and, in analogy with Eq. (A4),

\[
D_i = \frac{1}{n_i^2(\omega)} - \frac{1}{n_i^2(\omega)}. \tag{B3}
\]

By substituting the expression for \( \cos^2 \alpha \) as a function of \( n_p^2 \), as obtained from Eq. (5), into Eq. (B2), we transformed the square of Eq. (B1) into an equation involving two variables only, \( n_p^2 \) and \( n_i^2 \):

\[
D_i^2 n_i^4(\omega)(\omega_i^2 n_p^2 - \omega_i^2 n_s^2 + \omega_i^2 n_i^2)^4 + 4\omega_i^2 \omega_p^2 n_p^2 [\left( \frac{a}{n_p^2} + b \right) n_i^2 - \left( \frac{d}{n_p^2} + c \right) (\omega_i^2 n_p^2 - \omega_i^2 n_s^2 + \omega_i^2 n_i^2)^2] + 16 \left( \frac{g}{n_p^4} + \frac{h}{n_p^2 n_i^2} + l \right) \omega_i^4 n_i^4 \omega_i^4 n_i^4 - 32 \left( \frac{e}{n_p^2} + f \right) \times \omega_i^4 n_i^2 \omega_i^4 n_i^4 + 16n_i^4(\omega)\omega_i^4 \omega_i^4 n_i^4 = 0, \tag{B4}
\]

where

\[
a = 2n_i^2(\omega)D_i \left[ n_i^2(\omega)D_i + 2 \right],
b = -2n_i^2(\omega)D_i \left[ n_i^2(\omega)D_i + 2 \right] - 2n_i^2(\omega)D_i \left[ n_i^2(\omega)D_i + 1 \right],
\]

\[
c = -4 \frac{n_i^2(\omega)D_i}{n_i^2(\omega)} - 2n_i^4(\omega)D_i,
\]

\[
d = 4n_i^4(\omega)D_i,
\]

\[
e = -n_i^4(\omega)D_i,
\]

\[
f = n_i^4(\omega)D_i + n_i^2(\omega) + n_i^4(\omega)D_i, \frac{n_i^2(\omega)}{n_i^2(\omega)} D_i
\]

\[
g = n_i^4(\omega)D_i^2,
\]

\[
h = -2n_i^4(\omega)D_i^2 - 2n_i^2(\omega)D_i \left[ n_i^2(\omega)D_i + 1 \right] + \frac{n_i^4(\omega)D_i^2}{n_i^2(\omega)}, \tag{B5}
\]

Since, by substitution of Eq. (B4) into Eq. (9), \( n_i^2 \) can be expressed as a function of \( n_p^2 \), Eq. (B4) produces an eighth-degree polynomial equation for \( n_p^2 \). This equation can be numerically solved.

Condition (ii) is described by the set of Eqs. (1)–(6) and (7 ′) plus Eq. (10). Since this system is partially the same as that for condition (i), Eq. (B4) still holds for case (ii), where, instead of using Eq. (9), we substitute Eqs. (2)–(4) into Eq. (10) and obtain a second-degree equation for \( n_i^2 \) as a function of \( n_p^2 \). By substituting the solutions to this equation into the square of Eq. (B4), we get a 24th-degree polynomial equation for \( n_i^2 \) that can be numerically solved.

The physically acceptable solutions to either of the GV-matching conditions are fewer than those obtainable from the eight \( n_i^2 \) or 24 \( n_i^2 \) values above.

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### References