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Group-velocity-matched three wave mixing in birefringent crystals

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Abstract

We show that the combination of pulse-front slant, k-vector tilt, and crystal birefringence often permits exact matching of both phase and group velocities in three wave mixing in birefringent crystals. This makes possible more efficient mixing of short light pulses, and it permits efficient mixing of chirped or broad bandwidth light. We analyze this process and present examples.

Differences in the group velocities of the three interacting waves in a nonlinear crystal often limits the effective interaction length. For example, in mixing very short pulses, temporal walk off can stretch the pulses in time unless the crystal is very short. Efficient mixing with such short crystals requires high irradiances, but the irradiances are limited by higher order nonlinear effects such as intensity-dependent refractive index and two-photon absorption. Improved matching of the group velocities can alleviate this problem, allowing longer crystal and lower irradiances. Similarly, for high energy pulses, practical limits on crystal apertures mandate temporally stretching the pulses to reduce irradiances. For the resulting chirped pulses, temporal walk off restricts the chirp range unless the group velocities are well matched[1]. In addition to perfectly matching the group velocities of all three waves, it is sometimes useful to match two velocities, such as the signal and idler in parametric amplification, permitting broadband parametric amplification[2–5], or to arrange the velocities of two inputs to bracket the generated sum frequency pulse, giving pulse compression under

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suitable circumstances[6].

The two paramters that can be manipulated for group velocity adjustment of three fixed frequency pulses are the noncollinear phase matching angle and the pulse-front slant. Figure 1 shows an example. The pump propagation vector, k_p, is tilted by θ relative to the crystal's optic axis. This angle is dictated by phase matching for a signal tilt of δ relative to the pump. The corresponding idler angle, γ, must close the triangle of propagation vectors. The pulses are assumed to have parallel envelopes indicated by the heavy line slanted by ϕ relative to a normal to the pump propagation vector. Independent adjustment of δ and ϕ, while maintaining phase matching, allows flexible adjustment of the three group velocities. Because evaluation of simultaneous phase and group velocity matching can be tedious, we offer a computer program that computes noncollinear phase matching angles and the corresponding group velocities, using as inputs the nonlinear crystal, the polarization directions, the slant angle ϕ, and the wavelengths[7].

In previous work 5-10 fs pulses between 500 and 700 nm were created by parametric amplification of chirped signal light using a 150 fs, $\approx$390 nm, unchirped pump pulse[3,4]. The signal and idler group velocities were matched using noncollinear propagation with both pulse-fronts perpendicular to the signal k-vector. The pump group velocity differed from the signal and idler. They used type I mixing in a 1 mm thick, $\approx 32°$ cut, BBO crystal with a signal-to-pump angle of 3.7°. The amplified signal light was compressed after amplification to 5-10 fs. Riedle *et al.*[2] give an approximate general expression for the signal-to-pump angle required to group velocity match the signal and idler for type I mixing. Danielius *et al.*[8] pointed out that the combination of pulse-front slant and birefringent walk off can be used to adjust the group velocity of an extraordinary polarized wave in a birefringent crystal. They used this to set the group velocity of the pump midway between the signal and idler group velocities for collinear phase matching of type I mixing in BBO. For equal signal and idler wavelengths this gives perfect group velocity matching. What has not been exploited is that the combination of pulse-front slant and noncollinear phase matching provides great flexibility in adjusting the group velocities of the three waves. We examine the possibilities

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of this combination, with emphasis on exact group velocity matching of all three waves for arbitrary choices of wavelength.

We use the diagram in Fig. 2 to illustrate the calculation of the group velocity for a slanted pulse as measured along the $\hat{z}$ direction which is not collinear with the propagation vector that parallels v. Again the dark line represents the pulse front slanted at angle ϕ relative to a normal to $\hat{z}$. The vector v, parallel to the k-vector of the carrier wave of the pulse, represents the usual group velocity for a pulse with pulse front perpendicular to k. The birefringent walk off angle is ρ, so the pulse envelope propagates along v' which represents the ordinary group velocity along the Poynting vector. We find the group velocity along the $\hat{z}$ direction as follows:

$$v' = v/\cos(\rho) \tag{1}$$

$$h = v' \sin(\delta - \rho) \tag{2}$$

$$v_z = v' \cos(\delta - \rho) - h \tan(\phi) \tag{3}$$

$$v_z = v \frac{\cos(\delta - \rho) - \tan\phi \sin(\delta - \rho)}{\cos\rho}. \tag{4}$$

Using the small angle approximation for ρ, this can be written

$$v_z = v(\cos\delta - \tan\phi \sin\delta + \rho(\sin\delta + \tan\phi\cos\delta)) \tag{5}$$

This is the velocity at which the pulse front sweeps along the $\hat{z}$ axis.

Alternatively, we can derive v_z analytically. The group velocity along the $\hat{z}$ axis is given by

$$\frac{1}{v_z} = \frac{dk_z}{d\omega} = \frac{d(k\cos\delta)}{d\omega} = \frac{dk}{d\omega}\cos\delta - k\sin\delta\frac{d\delta}{d\omega}. \tag{6}$$

A slanted pulse front implies angular dispersion of the frequencies comprising the pulse. In the presence of birefringence, the refractive index is angle dependent so we must account for both the frequency and angle variation of k, so we rewrite the equation as

$$\frac{1}{v_z} = \left(\frac{\partial k}{\partial \omega} + \frac{\partial k}{\partial \delta}\frac{d\delta}{d\omega}\right)\cos\delta - k\sin\delta\frac{d\delta}{d\omega}. \tag{7}$$

Using the usual definitions of group velocity, v, and birefringent walk off, ρ, this can be rewritten

$$\frac{1}{v_z} = \left(\frac{1}{v} + k\rho\frac{d\delta}{d\omega}\right)\cos\delta - k\sin\delta\frac{d\delta}{d\omega}. \tag{8}$$

To evaluate $(d\delta/d\omega)$, we imagine creating a slanted pulse inside a birefringent crystal by diffracting an unslanted pulse off an embedded diffraction grating as shown in Fig. 2. A pulse with slant angle ψ relative to its k vector will be created if the diffraction angle is ψ. Diffraction must obey

$$k(\theta, \psi)\sin\psi = k_g \tag{9}$$

where k_g is the grating vector, $2\pi/d$, d being distance between grating lines. Differentiating with respect to ω gives

$$\left(\frac{\partial k}{\partial \psi}\frac{d\psi}{d\omega} + \frac{\partial k}{\partial \omega}\right)\sin\psi + k\cos\psi\frac{d\psi}{d\omega} = 0. \tag{10}$$

Again using the definitions of group velocity and walk off, we find

$$\frac{d\psi}{d\omega} = \frac{-1}{kv}\left(\frac{\sin\psi}{\rho\sin\psi + \cos\psi}\right). \tag{11}$$

This means that in Eq. (8) we can make the substitution

$$\frac{d\delta}{d\omega} = \frac{-1}{kv}\left(\frac{\sin\phi}{\rho\sin\phi + \cos\phi}\right), \tag{12}$$

yielding the same result as Eq. (5).

If we can find a set of angles (δ, γ, ϕ) that make v_z equal for all three pulses while also achieving phase matching, the pulses will stay overlapped in time as they propagate, although they will separate laterally due to birefringence and beam tilt. Obviously for large diameter beams this eliminates the problems associated with temporal walk off such as temporal broadening and reduced efficiency due to limited overlap distances. This should make possible more efficient mixing if the pulses have sufficient energy to permit large diameters. For the high pulse energies of terawatt systems, it is often necessary to stretch the pulses in time to keep beam diameters small enough to match available crystal sizes. The resulting chirped pulses are mixed and then compressed. Group velocity matching is ideal for this as it permits the mixing of pulses with arbitrary chirps. This can be seen by considering the phase mismatch with detuning of each wave from its carrier frequency. The phase mismatch along the $\hat{z}$-axis to first order in frequency shift is

$$\Delta k_z = \left(\frac{dk_z}{d\omega}\right)_p \Delta\omega_p - \left(\frac{dk_z}{d\omega}\right)_s \Delta\omega_s - \left(\frac{dk_z}{d\omega}\right)_i \Delta\omega_i \tag{13}$$

but if the group velocities along the $\hat{z}$-axis are all equal, this reduces to

$$\Delta k_z = \frac{1}{v_z}(\Delta\omega_p - \Delta\omega_s - \Delta\omega_i) \tag{14}$$

so the only requirement for maintaining phase matching is that the frequencies satisfy ($\omega_p = \omega_s + \omega_i$). Further, it is easy to show that if you choose a $\hat{z}'$ axis tilted relative to $\hat{z}$, group velocity matching along $\hat{z}$ implies group velocity matching along $\hat{z}'$. This means that transverse as well as longitudinal phase matching is maintained if the frequencies satisfy($\omega_p = \omega_s + \omega_i$). Dispersive elements such as prisms or gratings can be used to induce slanted pulse fronts for short pulses[9]. Using the same dispersion for chirped pulses sweeps the propagation angles in concert with the frequencies so phase matching is maintained throughout.

Pulse slant also contributes an anomalous group velocity dispersion[9] which combines with the usual group velocity dispersion to give a second order contribution to the phase mismatch of

$$\Delta k_z^{(2)} = \frac{1}{2}\left(\frac{d^2k_z}{d\omega^2}\right)_p (\Delta\omega_p)^2 - \frac{1}{2}\left(\frac{d^2k_z}{d\omega^2}\right)_s (\Delta\omega_s)^2 - \frac{1}{2}\left(\frac{d^2k_z}{d\omega^2}\right)_i (\Delta\omega_i)^2 \tag{15}$$

that limits the permissible pulse length or chirp. Starting with

$$\frac{d^2k_z}{d\omega^2} = \frac{d}{d\omega}\left(\frac{1}{v_z}\right) = \frac{-1}{v_z^2}\frac{dv_z}{d\omega} \tag{16}$$

and using Eq. 11 we find

$$\frac{d^2k_z}{d\omega^2} = -\frac{\text{GVD}}{vv_z} + \frac{1}{kvv_z}\left(\frac{\tan\phi}{1+\rho\tan\phi}\right)\left(\frac{\rho - \tan(\phi+\delta)}{1+\rho\tan(\phi+\delta)}\right), \tag{17}$$

where GVD is the ordinary group velocity dispersion along its propagation vector for an unslanted pulse

$$\text{GVD} = \frac{dv}{d\omega}. \tag{18}$$

To illustrate the flexibility of noncollinear phase matching combined with pulse-front slant, we used the SNLO function GVM to search for examples of group velocity matching for all three waves for the process (800 nm↔1400 nm + 1867 nm). In table I we show a few of the dozen plus successes.

We conclude that noncollinear mixing with slanted pulses gives great flexibility in adjusting the group velocities of the three interacting waves, including the possibility of exact group velocity for almost any set of wavelengths. This makes possible more efficient mixing of short or chirped pulses, with reduced influence from higher order processes of nonlinear refractive index and two-photon absorption. We developed general expressions for the effective group velocity and group velocity dispersion of slanted pulses in birefringent crystal and implemented them in SNLO[7] to eliminate the tedium of searching for experimental conditions that give a desired set of group velocities.

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FIGURES

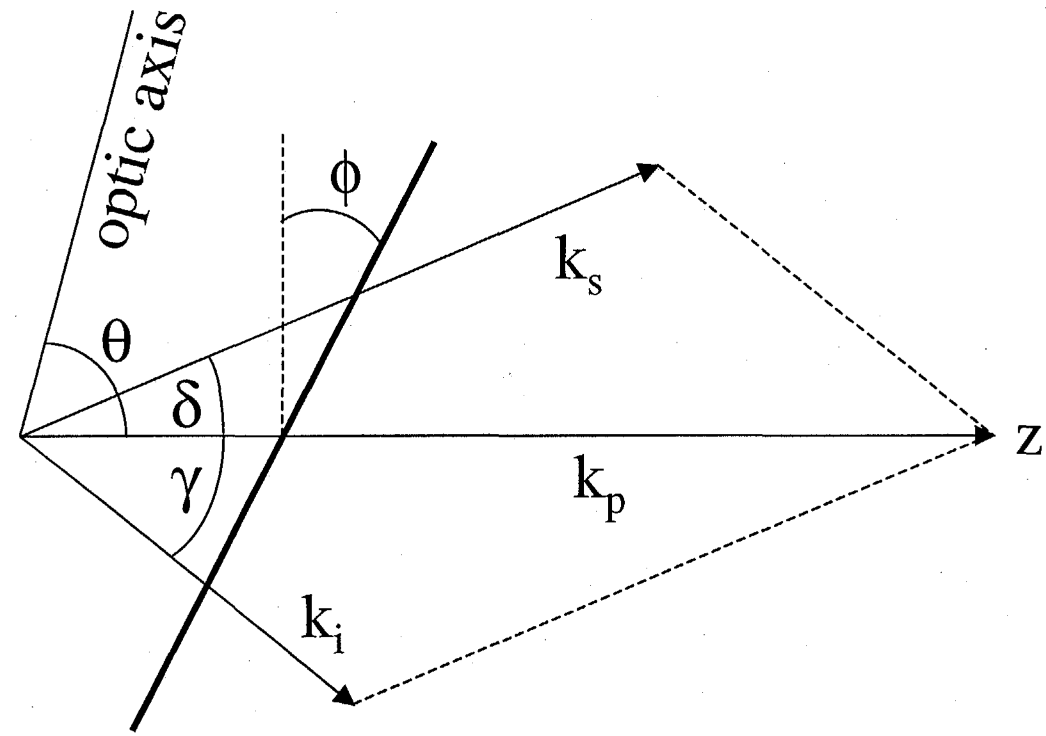
Fig. 1. Phase matching diagram for noncollinear mixing. The signal wave propagation vector k_s is tilted by angle δ with respect to the pump vector k_p; the idler wave propagation vector is tilted in the opposite direction by γ such that the vectors form a closed triangle in order to phase match. The pump propagation vector makes an angle of θ with the optic axis of the crystal. The envelope of each pulse is slanted by ϕ relative to a line normal to the k_p direction. For convenience we call the direction of k_p the $\hat{z}$ axis.

Fig. 2. Diagram for calculation of the group velocity of a slanted pulse along the $\hat{z}$-axis. The propagation vector is tilted by δ relative to the $\hat{z}$-axis and the usual group velocity along this direction is v. The birefringent walk off angle is ρ and the slant angle relative to the normal to the $\hat{z}$-axis is ϕ.

TABLES

Table 1. Examples of group-velocity-matched mixing of 800 nm, 1400 nm, and 1867 nm.

crystal (principal plane)	θ	δ	γ	polarizations (1867,1400,800 nm)	slant
BBO	35.2°	2.34°	1.80°	oee	-10.5°
BBO	28.95°	-1.8°	-1.32°	eoe	-7.1°
CLBO	45.7°	1.58°	1.2°	oee	-17.8°
KTP(YZ)	74.5°	6.82°	5.30°	eoo	11.77°
$LiIO_3$	26.4°	7.2°	5.37°	ooe	-0.19°


optic axis
θ
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ϕ
k_s
k_p
k_i
z

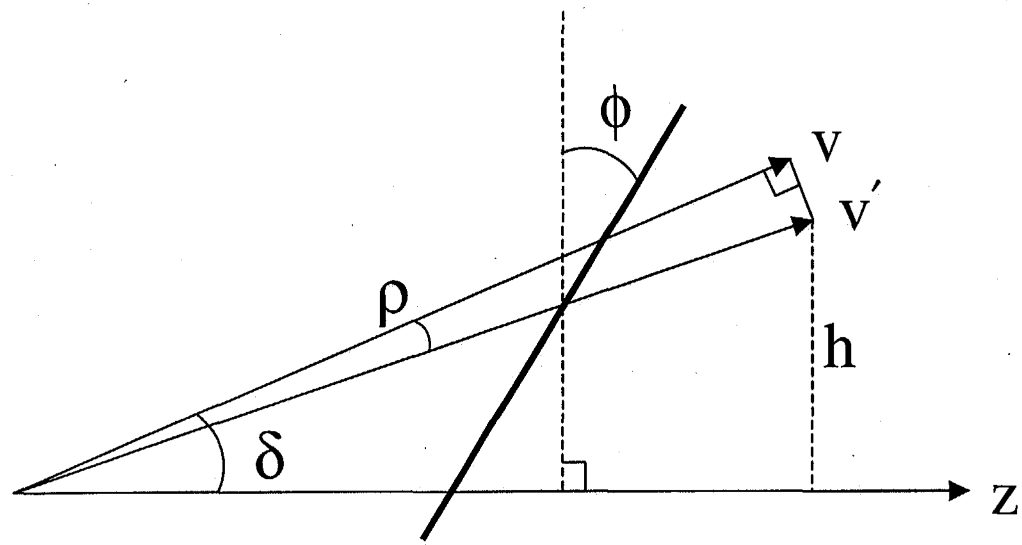

Fig 2