Group-velocity-matched three-wave mixing in birefringent crystals

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It is shown that the combination of pulse-front slant, k-vector tilt, and crystal birefringence often permits exact matching of both phase and group velocities for three-wave mixing in birefringent crystals. This exact match makes possible more-efficient mixing of short light pulses, and it permits efficient mixing of chirped or broadband light. I analyze this process and present examples. © 2001 Optical Society of America

OCIS codes: 190.4410, 190.7110.

Differences among the group velocities of three interacting waves in a nonlinear crystal often limit the effective interaction length. For example, in mixing short pulses, temporal walk-off can exceed the pulse durations unless the crystal is also short. Efficient mixing with such short crystals requires high irradiances, but irradiances may be limited by higher-order nonlinear effects such as an intensity-dependent refractive index and two-photon absorption. Improved group-velocity matching would permit longer crystals and lower irradiances. Similarly, for high-energy pulses, practical limits on crystal apertures mandate temporally stretching the pulses to reduce irradiances. For the resultant chirped pulses, temporal walk-off restricts the chirp range unless the group velocities are well matched. In addition to matching the group velocities of all three waves, it is sometimes useful to match only two velocities, such as the signal and the idler in parametric amplification, permitting broadband parametric amplification,[1-4] or to arrange the velocities of the two input pulses to bracket that of the generated sum-frequency pulse, giving pulse compression under suitable conditions.[5]

The two parameters that can be manipulated to adjust the group velocities of three fixed-frequency pulses are the noncollinear phase matching angles and the pulse-front slant. Figure 1 shows an example of this. The pump propagation vector, $\mathbf{k}^p$, 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. All three pulses are assumed to have slanted but parallel envelopes, or pulse fronts, indicated by the thick bold line slanted by ϕ relative to a normal to $\mathbf{k}^p$. If the pulses are to stay temporally overlapped as they propagate, they must have the same group velocity as measured along a common axis, which was chosen for convenience to be $\mathbf{k}^p$. Independent adjustment of δ and ϕ while phase matching is maintained permits flexible adjustment of the three group velocities.

Previously, 5–10-fs pulses from 500 to 700 nm were created by parametric amplification of chirped signal light with a 150-fs, ≈390-nm unchirped pump pulse.[2,3] The signal and idler group velocities were matched by noncollinear propagation with both pulse fronts perpendicular to $\mathbf{k}^s$, but the pump's group velocity varied. The pulses used type I mixing in a 1-mm thick, ≈32°-cut, β-barium borate crystal with $\delta = 3.7°$. The amplified signal light was compressed to 5–10 fs. Riedle *et al.*[1] give an approximate general expression for the signal-to-pump angle required for matching signal and idler group velocities for type I mixing. Danielius *et al.*[6] pointed out that the combination of pulse-front slant and birefringent walk-off can be used to adjust the group velocity of an extraordinarily polarized wave. They used this property to set the group velocity of the pump midway between those of the signal and the idler for collinearly phase-matched type I mixing in β-barium borate. For equal signal and idler wavelengths this technique can achieve perfect group-velocity matching. What has not been exploited is the combination of pulse-front slant and noncollinear phase matching that provides considerable flexibility in adjusting the group velocities of the three waves. Here we examine the possible uses of this combination, with emphasis on exact group-velocity matching of all three waves for arbitrary choices of wavelength.

Figure 2 illustrates my calculation of the group velocity along $\hat{z}$ ($= \hat{k}^p$) for a slanted pulse. This is the velocity v_z at which point a sweeps along $\hat{z}$. Again
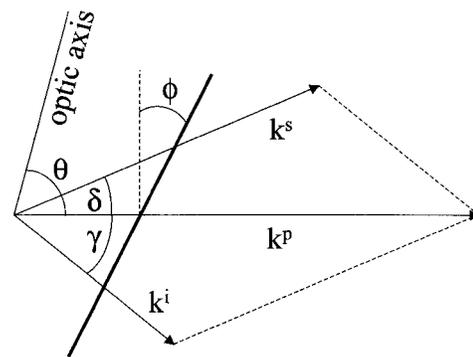
Fig. 1. Phase-matching diagram of noncollinear mixing. Vectors $\mathbf{k}^s$, $\mathbf{k}^i$, and $\mathbf{k}^p$ are the signal, the idler, and the pump propagation vectors, respectively. The thick bold line represents the parallel envelopes of all three pulses.
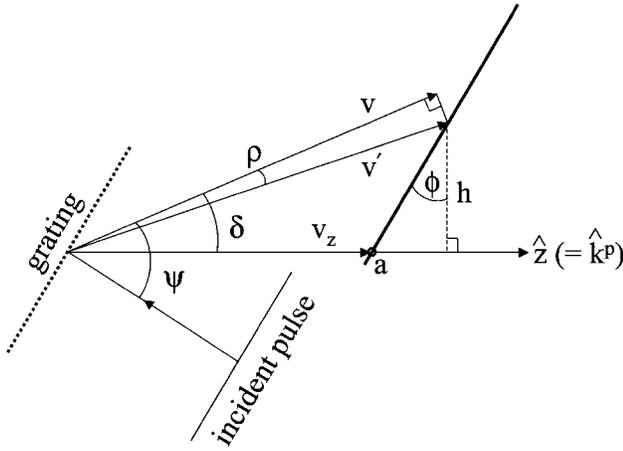
Fig. 2. Diagram for calculation of the group velocity of a slanted pulse along $\hat{z}$. The propagation vector is tilted by δ relative to $\hat{z}$, and v is the group velocity of an unslanted pulse. The birefringent walk-off angle is ρ, and ϕ is the slant angle of the pulse front relative to the normal to $\hat{z}$. The velocity at which point a sweeps along the $\hat{z}$ axis is v_z. The grating is shown for heuristic purposes only.

the thick bold line represents the slanted pulse front. Vector **v**, parallel to the k vector of the pulse's carrier wave, represents the group velocity of a pulse whose pulse front is perpendicular to **k**. If the pulse has extraordinary polarization, its Poynting vector is tilted by ρ relative to **k**, so the group velocity of an unslanted pulse becomes **v′**. We find v_z as follows:

$$v' = v/\cos \rho, \qquad h = v' \sin(\delta - \rho), \qquad (1)$$

$$v_z = v' \cos(\delta - \rho) - h \tan \phi$$

$$= v \frac{\cos(\delta - \rho) - \tan \phi \sin(\delta - \rho)}{\cos \rho} . \qquad (2)$$

Using the small angle approximation for ρ, we can write v_z as

$$v_z = v[\cos \delta - \tan \phi \sin \delta$$

$$+ \rho(\sin \delta + \tan \phi \cos \delta)]. \qquad (3)$$

Alternatively, we can derive v_z analytically starting with

$$\frac{1}{v_z} = \frac{\mathrm{d}k_z}{\mathrm{d}\omega} = \frac{\mathrm{d}(k \cos \delta)}{\mathrm{d}\omega}$$

$$= \frac{\mathrm{d}k}{\mathrm{d}\omega} \cos \delta - k \sin \delta \frac{\mathrm{d}\delta}{\mathrm{d}\omega} . \qquad (4)$$

A slanted pulse front implies angular dispersion of the frequencies that compose the pulse. In the presence of birefringence, the refractive index is angle dependent, so we must account for both the frequency and the angle variation of k. We rewrite Eq. (4) as

$$\frac{1}{v_z} = \left(\frac{\partial k}{\partial \omega} + \frac{\partial k}{\partial \delta} \frac{\mathrm{d}\delta}{\mathrm{d}\omega} \right) \cos \delta - k \sin \delta \frac{\mathrm{d}\delta}{\mathrm{d}\omega} . \qquad (5)$$

Using the usual definitions of group velocity $v = \partial \omega / \partial k$ and birefrigent walk-off $\rho = (1/n) \partial n / \partial \delta$, we can rewrite Eq. (5) as

$$\frac{1}{v_z} = \left(\frac{1}{v} + k\rho \frac{\mathrm{d}\delta}{\mathrm{d}\omega} \right) \cos \delta - k \sin \delta \frac{\mathrm{d}\delta}{\mathrm{d}\omega} . \qquad (6)$$

To evaluate $(\mathrm{d}\delta/\mathrm{d}\omega)$ we imagine creating a slanted pulse inside a birefringent crystal by diffracting an unslanted pulse off an embedded diffraction grating aligned parallel with the incident pulse front, 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

$$k(\omega, \psi) \sin \psi = k_g , \qquad (7)$$

where k_g is the grating vector. Differentiating with respect to ω gives

$$\left(\frac{\partial k}{\partial \psi} \frac{\mathrm{d}\psi}{\mathrm{d}\omega} + \frac{\partial k}{\partial \omega} \right) \sin \psi + k \cos \psi \frac{\mathrm{d}\psi}{\mathrm{d}\omega} = 0 . \qquad (8)$$

Again using the definitions of group velocity and walk-off, Eq. (8) gives

$$\frac{\mathrm{d}\psi}{\mathrm{d}\omega} = \frac{-1}{kv} \left(\frac{\sin \psi}{\rho \sin \psi + \cos \psi} \right) . \qquad (9)$$

Using the relations $\mathrm{d}\delta/\mathrm{d}\omega = \mathrm{d}\psi/\mathrm{d}\omega$ and $\psi = \phi + \delta$ gives

$$\frac{\mathrm{d}\delta}{\mathrm{d}\omega} = \frac{-1}{kv} \left[\frac{\sin(\phi + \delta)}{\rho \sin(\phi + \delta) + \cos(\phi + \delta)} \right] . \qquad (10)$$

Substituting Eq. (10) into Eq. (6) and simplifying yields Eq. (3).

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 because of birefringence and noncollinearity. For large-diameter beams this lateral walk-off may be insignificant, so this method of group-velocity matching can minimize the problems associated with temporal walk-off, making possible longer interaction lengths and more-efficient mixing with little pulse broadening.

For the high pulse energies of terawatt systems it is often necessary to stretch the pulse in time to keep beam diameters small enough to match available crystal apertures. The resultant chirped pulses are mixed and then compressed. Group-velocity matching is ideal for this because it permits the mixing of pulses with arbitrary chirps, as one can see 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

$$\Delta k_z^{(1)} = \frac{\mathrm{d}k_z^p}{\mathrm{d}\omega} \Delta \omega_p - \frac{\mathrm{d}k_z^s}{\mathrm{d}\omega} \Delta \omega_s - \frac{\mathrm{d}k_z^i}{\mathrm{d}\omega} \Delta \omega_i , \qquad (11)$$

but, if the group velocities along $\hat{z}$ are all equal to v_z, Eq. (11) reduces to

$$\Delta k_z^{(1)} = \frac{1}{v_z} (\Delta \omega_p - \Delta \omega_s - \Delta \omega_i) , \qquad (12)$$

Table 1. Examples of Group-Velocity-Matched Mixing of Three Interacting Waves at 800, 1400, and 1867 nm

Crystal[a]	θ (°)	δ (°)	γ (°)	Polarization[b]	Slant (°)
β-BaB$_2$O$_4$	35.2	2.34	1.80	oee	-10.5
β-BaB$_2$O$_4$	28.95	-1.8	-1.32	eoe	-7.1
CeLiB$_6$O$_{16}$	45.7	1.58	1.2	oee	-17.8
KTiOPO$_4$	74.5	6.82	5.30	eoo	11.77
LiIO$_3$	26.4	7.2	5.37	ooe	-0.19

[a]Principal plane.
[b]1867, 1400, and 800 nm.

so the only requirement for maintaining phase matching is that the frequencies satisfy ($\Delta\omega_p = \Delta\omega_s + \Delta\omega_i$), which is automatically imposed by frequency mixing. Further, it is easy to show that if one chooses a $\hat{z}'$ axis tilted relative to $\hat{z}$, group-velocity matching along $\hat{z}$ implies group-velocity matching along $\hat{z}'$, so transverse as well as longitudinal phase matching will be achieved. Dispersive elements such as prisms and gratings can be used to induce the required pulse-front slant for short pulses.[7] Using the same prism or grating for chirped pulses sweeps the propagation angles in concert with the frequencies in such a way that phase matching is maintained throughout the stretched pulse.[8]

Pulse slant also contributes a group-velocity dispersion[7] that combines with the usual group-velocity dispersion to make a second-order contribution to the phase mismatch in the $\hat{z}$ direction:

$$\Delta k_z^{(2)} = \frac{1}{2}\frac{\mathrm{d}^2 k_z{}^p}{\mathrm{d}\omega^2}(\Delta\omega_p)^2 - \frac{1}{2}\frac{\mathrm{d}^2 k_z{}^s}{\mathrm{d}\omega^2}(\Delta\omega_s)^2$$
$$- \frac{1}{2}\frac{\mathrm{d}^2 k_z{}^i}{\mathrm{d}\omega^2}(\Delta\omega_i)^2. \tag{13}$$

If group-velocity matching is achieved, this group-velocity dispersion will limit the permissible crystal length or chirp range. Starting with

$$\frac{\mathrm{d}^2 k_z}{\mathrm{d}\omega^2} = \frac{\mathrm{d}}{\mathrm{d}\omega}\left(\frac{1}{v_z}\right) = \frac{-1}{v_z{}^2}\frac{\mathrm{d}v_z}{\mathrm{d}\omega} \tag{14}$$

and using Eqs. (3) and (10), we find that

$$\frac{\mathrm{d}^2 k_z}{\mathrm{d}\omega^2} = -\frac{\mathrm{GVD}}{v v_z} + \frac{1}{k v v_z}\left[\frac{\tan(\phi + \delta)}{1 + \rho\,\tan(\phi + \delta)}\right]$$
$$\times\left[\frac{\rho - \tan(\phi + \delta)}{1 + \rho\,\tan(\phi + \delta)}\right], \tag{15}$$

where GVD $= \mathrm{d}v/\mathrm{d}\omega$, the ordinary group-velocity dispersion along its propagation vector for an unslanted pulse.

Because the calculations outlined above are tedious, I offer a public-domain computer program, function GVM within the nonlinear optics software SNLO,[9] which computes noncollinear phase-matching angles and group velocities. To illustrate the versatility of this method of group-velocity matching, I used GVM to search for examples of group-velocity matching of all three waves for (800 nm $\leftrightarrow$ 1400 nm + 1867 nm). Table 1 lists some of the dozen or more successful group-velocity matching conditions.

We conclude that noncollinear mixing with slanted pulses provides flexibility in adjusting the group velocities of three interacting waves, including the possibility of exact group-velocity matching for a wide range of wavelengths. This makes possible more-efficient mixing of short or chirped pulses, with reduced influence from higher-order nonlinear processes. General expressions for the effective group velocity and group-velocity dispersion of slanted pulses in birefringent crystal have been developed and implemented in SNLO[9] to expedite the search for experimental conditions that give a desired set of group velocities.

References

1. E. Riedle, M. Beutter, S. Lochbrunner, J. Piel, S. Schenkl, S. Sporlein, and W. Zinth, Appl. Phys. B **71,** 457 (2000).
2. A. Shirakawa, I. Sakane, M. Takasaka, and T. Kobayashi, Appl. Phys. Lett. **74,** 2268 (1999).
3. G. Cerullo, M. Nisoli, S. Stagira, S. De Silvestri, G. Tempea, F. Krausz, and K. Ferencz, Appl. Phys. B Suppl. **70,** S253 (2000).
4. A. Andreoni and M. Bondani, Appl. Opt. **37,** 2414 (1998).
5. A. Dubietis, G. Valiulis, R. Danielius, and A. Piskarskas, Pure Appl. Opt. **7,** 271 (1998).
6. R. Danielius, A. Piskarskas, P. Di Trapani, A. Andreoni, C. Solcia, and P. Foggi, Opt. Lett. **21,** 973 (1996).
7. J.-C. Diels and W. Rudolph, *Ultrashort Laser Pulse Phenomena* (Academic, New York, 1996).
8. B. A. Richman, S. E. Bisson, R. Trebino, E. Sidick, and A. Jacobson, Opt. Lett. **23,** 497 (1998).
9. Function GVM in SNLO versions 22 and higher. SNLO nonlinear optics software is available from the author as a free download at http://www.sandia.gov/imrl/XWEB1128/xxtal.htm.