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Large Phase Shifts due to the $\chi^{(2)}$ Cascading Nonlinearity in Large Walk-off and Loss Regimes in Semiconductors and Other Dispersive Materials

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The nonlinear phase shift associated with $\chi^{(2)}$ cascading was studied for ultrashort pulses in regimes with large temporal walk-offs between the fundamental and the $\chi^{(2)}$-generated second-harmonic (SH) pulses, a situation typical of semiconductors. Despite the small overlap between the fundamental and SH pulses after they walk-off each other, a nonlinear phase shift was numerically observed to occur at a rate comparable to that before they walk-off. A $\pi/2$ phase shift was demonstrated even in a waveguide 15 times longer than the walk-off length.

KEYWORDS: cascading nonlinearity, nonlieanr phase shift, temporal walk-off, dispersion, semiconductor

The $\chi^{(2)}$ cascading nonlinearity[1,2] is recently attracting new attention for applications as diverse as ultrafast switching,[3] optical transistor action,[4] and one- and two-dimensional spatial solitons.[5–7] One advantage of this off-resonant process with $\chi^{(2)}$ materials is that it requires a relatively small light intensity for inducing a particular nonlinear phase shift when compared with conventional $\chi^{(3)}$ processes. One limitation that has previously been believed to be serious is the tempral walk-off which occurs between fundamental and second-harmonic (SH) pulses in semiconductors and other dispersive media. For example, the maximum propagation length used in early experiments was limited to 1.9 times the walk-off length ($L_{\text{walk-off}}$),[8] because the fundamental pulse was assumed to no longer interact with the SH pulse when the fundamental pulse walks off beyond the SH pulse. Because they exhibit very short walk-off lengths due to their strong refractive-index dispersion, semiconductors are apparently unsuitable for cascading, despite their large nonlinearities ($d_{14}^{(2)} = 90$–$130\,\text{pm/V}$, that is, $\chi_{xyz}^{(2)} = 4$–6×10^{-7} esu for GaAs, ref. 9) and their potential for integration with light sources and detectors. In fact, the cascading phase shift in strong walk-off regimes has never been investigated, either experimentally or theoretically. To the contrary of currently held opinion mentioned above, this paper shows that a significant cascading phase shift occurs even in a strong walk-off regime, based on numerical simulations. As an example pertinent to applications, the operating conditions for a semiconductor-based Mach-Zehnder switch was estimated.

The starting point was the usual coupled-mode equations,[1,8)]

$$\frac{\partial}{\partial z}u_\omega + k'_\omega \frac{\partial}{\partial t}u_\omega = -i\Gamma u_{2\omega}u_\omega^* \exp[-\mathrm{i}\Delta kz], \quad (1)$$

$$\frac{\partial}{\partial z}u_{2\omega} + k'_{2\omega} \frac{\partial}{\partial t}u_{2\omega} = -i\Gamma u_\omega^2 \exp[\mathrm{i}\Delta kz], \quad (2)$$

where u_ω and $u_{2\omega}$ are the fundamental and SH field amplitudes normalized as, $E_\omega(z, t) = u_\omega(z, t) \cdot E_\omega(z = 0, t = 0)$ and $E_{2\omega}(z, t) = u_{2\omega}(z, t) \cdot E_\omega(z = 0, t = 0)$. The walk-off terms are included via the group velocities k'_ω and $k'_{2\omega}$, while the group velocity dispersion within each pulse's spectrum is neglected. The phase mismatch Δk is defined as $\Delta k = k_{2\omega} - k_\omega$. The normalized input field Γ is defined as,

$$\Gamma = \frac{\omega \cdot d_{\text{eff}} : |E_\omega(z = 0, t = 0)|}{c\sqrt{n_\omega n_{2\omega}}},$$

where d_{eff} is the effective second-order susceptibility and c is the light velocity. For an input pulse of width T_0 and a total propagation distance L, the coupled-mode equations in a dimensionless form used in this work are,

$$\frac{\partial}{\partial \xi}u_w = -i(\Gamma L)u_{2w}u_w^* \exp[-\mathrm{i}(\Delta kL)\xi], \quad (3)$$

$$\frac{\partial}{\partial \xi}u_{2w} + \frac{(k'_{2\omega} - k'_\omega)\cdot L}{T_0}\frac{\partial}{\partial \tau}u_{2w} = -i(\Gamma L)u_w^2 \exp[i(\Delta kL)\xi]. \quad (4)$$

Here, z is normalized as $\xi = z/L$. A moving reference frame $\tau = (t - k'_\omega z)/T_0$ was used so that $u_w(\xi, \tau = 0)$ indicates the fundamental pulse's peak amplitude throughout $0 \le z \le L$, while $u_{2w}(\xi, \tau = 0)$ indicates the SH field amplitude at the fundamental pulse's peak position. The pulse walk-off factor $(k'_{2\omega} - k'_\omega)L/T_0$ in eq. (4) is simplified to $L/L_{\text{walk-off}}$, when using the walk-off length defined as $L_{\text{walk-off}} = T_0/(k'_{2\omega} - k'_\omega)$. Thus, the independent variables for this set of equations are the cumulative phase mismatch ΔkL, the normalized input power $(\Gamma L)^2$, and the pulse walk-off $L/L_{\text{walk-off}}$.

Figure 1 shows a sample simulation of the evolution of the fundamental pulse interacting with a SH pulse created by $\chi^{(2)}$ in a regime where the total propagation distance (L) is $15L_{\text{walk-off}}$. Because the SH intensity right at the fundamental pulse's peak is very weak in such a large walk-off regime, the cascading interaction could be reasonably assumed to *not* be significant when compared with the evolution without walk-off (Fig. 2). To the contrary, the fundamental pulse gains a $\pi/2$ nonlinear phase shift according to the calculations, as shown at $L/L_{\text{walk-off}} = 15$ in Fig. 3. The fundamental throughput is as large as 0.33, even though the fundamental pulse energy converted to SH effectively never returns to the fundamental by down-conversion. Here, the cumulative phase mismatch ΔkL was assumed to be π^2, and the normalized input power $(\Gamma L)^2$ was 120. This geometry gives a net phase shift of about 1.2π in the limit of no

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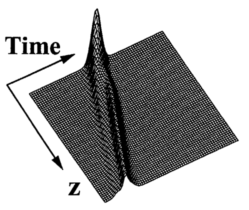
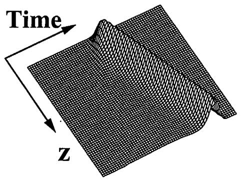
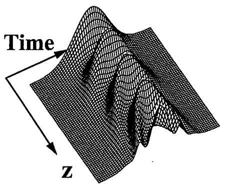


(a) Fundamental Pulse



(b) Second-Harmonic Pulse

Fig. 1. Simulated evolution of the fundamental (a) and second-harmonic (SH) (b) pulses, in a large walk-off regime ($L = 15L_{\text{walk-off}}$). The fundamental pulse acquires a $\pi/2$ nonlinear phase shift, while that in the no-walk-off limit is approximately 1.2π. $\Delta\kappa L = \pi^2$ and $(\Gamma L)^2 = 120$ are assumed. The time scale ranges from $-11T_0$ to $+11T_0$, where T_0 is the initial pulse width. The z scale ranges from 0 to L.



Fundamental Pulse

Fig. 2. Simulated evolution of the fundamental pulse in the no-walk-off limit, where the SH pulse co-propagates. The same parameters (except for walk-off) are assumed. The time scale ranges from $-2.7T_0$ to $+2.7T_0$, where the pulse break-up as well as the oscillation (due to down conversion) are clearly shown.

walk-off ($L/L_{\text{walk-off}} = 0$). Another set of results obtained for $\Delta kL = 10\pi$ and $(\Gamma L)^2 = 400$ also showed a large nonlinear phase shift.

When a 1-psec 1.55-μm pulse propagates in a typical semiconductor waveguide, $L = 15L_{\text{walk-off}}$ corresponds typically to 4.5mm. $(\Gamma L)^2 = 120$ implies a 20 W input power, assuming an effective second-order susceptibility $d_{ij}^{(2)}$ of 100 pm/V for the waveguide.[10] This peak input power would be realistic for practical applications. Figure 1 also shows that the walk-off almost completely eliminates the pulse "break-up" which normally ocurrs for such relatively small phase mismatchs ($\Delta kL = \pi^2$) as indicated in Fig. 2. This would be a distinct advantage for applications.

An underlying key feature which leads to the large phase shift was found to be the evolution of phase relationship between the fundamental and SH fields. After re-defining the fundamental and SH amplitudes as $u_\omega = |u_\omega| \exp[\mathrm{i}\Phi_\omega]$ and $u_{2\omega} = |u_{2\omega}| \exp[\mathrm{i}\Phi_{2\omega}]$, we derived a set of equations from the down-conversion eq. (eq. (3)),

$$\frac{\partial}{\partial \xi}\Phi_\omega = -(\Gamma L)\cdot|u_{2\omega}|\cdot\cos[2\Phi_\omega - \Phi_{2\omega} + \Delta kL\xi], \quad (5)$$

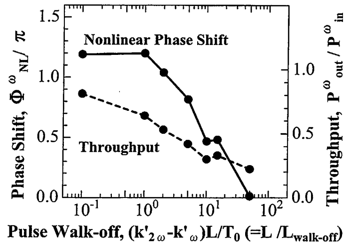


Fig. 3. The pulse-walk-off dependencies of the nonlinear phase shift and the throughput of the fundamental pulse ($|u_\omega|^2$). The propagation distance L is kept constant. A $\pi/2$ shift occurs even at $(k'_{2\omega} - k'_\omega)L/T_0 (= L/L_{\text{walk-off}}) = 15$ ($\Delta\kappa L = \pi^2$ and $(\Gamma L)^2 = 120$).

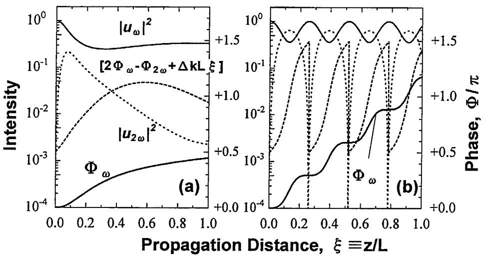


Fig. 4. Evolution of the nonlinear phase shift $\Phi_\omega(\xi, \tau = 0)$ together with the fundamental-pulse (solid line) and second-harmonic-pulse (dotted line) intensities at the fundamental pulse's peak position ($\tau = 0$). (a) in the walk-off regime $[L/L_{\text{walk-off}} = 15, \Delta\kappa L = \pi^2, (\Gamma L)^2 = 120]$. The relative phase shift (dashed line) remains near $+\pi$ from $z = L/3$ through $z = L$. (b) in the no-walk-off limit $[L/L_{\text{walk-off}} = 0, \Delta\kappa L = \pi^2, (\Gamma L)^2 = 120]$ for comparison.

and

$$\frac{\partial}{\partial \xi}|u_\omega| = -(\Gamma L)\cdot|u_{2\omega}|\cdot|u_\omega|\cdot\sin[2\Phi_\omega - \Phi_{2\omega} + \Delta kL\xi]. \quad (6)$$

Equation (5) is responsible for the nonlinear phase shift of the fundamental pulse at its peak $\Phi_\omega(\xi, \tau = 0)$ (henceforth, all variables are discussed at the fundamental pulse's peak position, $\tau = 0$). After the SH pulse walks off beyond the fundamental pulse in the region $L/3 < z < L$, the cosine factor $\cos[2\Phi_\omega(\xi, \tau = 0) - \Phi_{2\omega}(\xi, \tau = 0) + \Delta kL\xi]$ was found to stay close to unity because the relative phase $[2\Phi_\omega(\xi, \tau = 0) - \Phi_{2\omega}(\xi, \tau = 0) + \Delta kL\xi]$ remains approximately $+\pi$ (Fig. 4(a)). This feature was confirmed clearly in a wide range of conditions ($L/L_{\text{walk-off}} = 3$–$20$ for $[\Delta\kappa L = \pi^2, (\Gamma L)^2 = 120]$, $L/L_{\text{walk-off}} = 2$–$50$ for $[\Delta\kappa L = 10\pi, (\Gamma L)^2 = 400]$, etc.). Thus, the nonlinear phase shift occurs continuously, despite the lack of strong down conversion. In contrast to this behaviour, without the walk-off $[L/L_{\text{walk-off}} = 0]$ the relative phase keeps changing between $+\pi/2$ and $+3\pi/2$ and therefore a significant phase shift occurs only when the relative phase crosses $+\pi$ (Fig. 4(b)). In fact, the fundamental pulse in the walk-off regime in Fig. 4(a) acquired one half of its total phase shift in propagating from $z = L/3$ ($= 5L_{\text{walk-off}}$) to L, although the SH intensities at the fundamental pulse's peak ($|E_{2\omega}(\tau = 0)|^2$) at $z = L/3$ and $z = L$ were one and two orders of magnitude smaller respectively than the SH peak intensity near $z = 0$. Even with this small SH field co-propagating

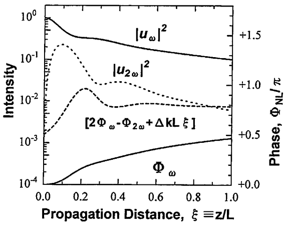


Fig. 5. Evolution of the phases and intensities in a no-walk-off regime, but with large second-harmonic loss. A normalized loss ($\alpha_{2\omega}L$) of 20 was assumed. Despite the small second-harmonic intensity from $z = L/3$ through L, a $\pi/2$ shift occurs again. The relative phase shift remains near $+\pi$, in a way similar to that in the walk-off regime.

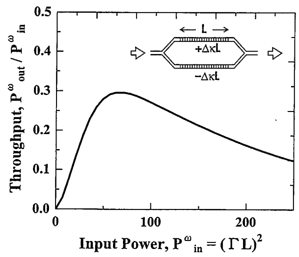


Fig. 6. The fundamental-pulse throughput for a Mach-Zehnder-interferometer switch (inset), in the walk-off regime ($L/L_{\text{walk-off}} = 15$). The hatched regions identify the two cascading regions which are biased to different sides of the SHG phase matching condition ($\Delta\kappa L = \pm\pi^2$).

with the fundamental pulse, the phase shift occurred at a rate comparable to that before they walk-off.[11] This is a consequence of the increase in the cosine factor which partly cancels the decrease in the $|E_{2\omega}|$ (*not* $|E_{2\omega}|^2$) factor in eq. (5). Similarly, the relative phase in the sine term in eq. (6) suppresses the loss of fundamental pulse energy. This is consistent with the fact that minimal SH is generated during the propagation except at the very beginning stage ($z < L/3$), as previously seen in Fig. 1.

A large phase shift was also observed in a manner similar to that just discussed for the walk-off regime, when a large loss for the second-harmonic light ($\alpha_{2\omega}$) was assumed in the waveguide. The SHG differential equation,

$$\frac{\partial}{\partial\xi}u_{2w} + \frac{\alpha_{2\omega}L}{2}u_{2w} = -i(\Gamma L)\cdot u_w^2 \exp[\mathrm{i}(\Delta kL)\xi], \quad (7)$$

was used in place of eq. (4). The normalized absorption $\alpha_{2\omega}L$ in Fig. 5 was chosen such that $|u_{2\omega}|^2$ at $z = L/3$ is one order of magnitude smaller than the SH peak intensity, as in Fig. 4(a). The absorption thus chosen was $\alpha_{2\omega}L = 20$, which means that the transmittance for second-harmonic light intensity of this waveguide is only $\exp(-20) = 2 \times 10^{-9}$. Despite this large absorption, Fig. 5 shows that a large nonlinear phase shift of $\pi/2$ still occurs. The relative phase was approximately $+\pi$ again, in a manner similar to that in the walk-off regime. Note that, both in the walk-off regime and in the loss regime the SH field at the fundamental pulse's peak is being "lost" along the propagation path by the walk-off and by the absorption respectively. These results suggest that a loss of the SH field "pins" the relative phase near $+\pi$, which leads to a continuous phase shift that suppresses the energy exchange between the fundamental and harmonic. This feature which covers both the walk-off and loss regimes is different from any of the results reported by DeSalvo *et al.*[1] or the cases discussed in the recent paper of Kobyakov and Lederer[12] in which they reviewed the theoretical work published to date. From the perspective of the generic type of system of equations involved, the two casess discussed in the present work are dissipative in nature. Previously, cascading nonlinearities in a dissipative system were rarely discussed, to the best of author's knowledge, except for the pioneering work by Armstrong *et al.*[13] Their focus, however, was on an exact solution for the special case, $\alpha_{2\omega} = \alpha_\omega$.

Finally, the feasibility of a Mach-Zehnder switch[14] was confirmed for the cascading conditions in the walk-off regime (Fig. 6). A pulse in each of the two cascading regions, which are biased to different sides of the phase-matching condition ($\Delta kL = \pm\pi^2$), acquires a $\pi/2$ shift within the 15 walk-off length distance. The switch-on curve is smooth, and the switch-on pulse shape exhbits no pulse break-up.

In summary, we have shown that $\pi/2$ phase shifts can be obtained both in a walk-off regime ($L/L_{\text{walk-off}} = 15$) and in a high harmonic-loss regime ($\alpha_{2\omega}L = 20$). These results should broaden the choice of $\chi^{(2)}$ materials that can be used for cascading, and also allow operation with ultrafast pulses.

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