Spectral phase correlation of coded femtosecond pulses by second-harmonic generation in thick nonlinear crystals

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We demonstrate a novel all-optical scheme for measuring the correlation of spectrally phase-coded ultrashort optical waveforms that uses second-harmonic generation (SHG) in long, periodically poled lithium niobate crystals. The SHG yield can be controlled over a range of ~30 dB, depending on the correlation of the applied phase codes. Such a spectral phase correlator has applications for ultrashort-pulse optical code-division multiple-access networking and could serve as a nonlinear optical but classical analog for certain schemes for coherent quantum control of multiphoton processes. © 2000 Optical Society of America

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Using ultrashort pulses opens new dimensions in studying second-harmonic generation (SHG), one of the earliest nonlinear optical phenomena discovered.[1] The second-harmonic (SH) signals can be influenced by the broad and sometimes complex spectral structure of the excitation pulses, besides their high peak intensity. Narrowing of the SHG spectrum was previously demonstrated by the use of simple spectral amplitude filters in conjunction with chirped femtosecond pulses.[2] The development of the pulse-shaping techniques[3–5] now permits high-accuracy modulation of the spectral amplitude and phase of ultrashort pulses. Here we study SHG by using femtosecond pulses that are spectrally phase coded with binary code sequences taken from communication theory and demonstrate the ability to control the SHG yield over a 3-order-of-magnitude range, depending on the code structure. Our results show that, under conditions of large group-velocity mismatch (GVM),[6] SHG in the perturbative limit could serve as a nonlinear optical spectral correlator. Our results are relevant both for control of quantum multiphoton processes[7] and for recognition of specially phase coded femtosecond waveforms in a proposed optical code-division multiple-access scheme.[8–10]

In ultrashort-pulse SHG, each SH spectral component results from the addition of different pairs of fundamental frequency components. When intrapulse group-velocity dispersion and pump depletion are negligible, the SH spectrum can be described by[11]

$$A_2(\Omega) = \int_0^\infty A_1(\Omega/2 + \Omega')A_1(\Omega/2 - \Omega')\mathrm{d}\Omega' \times D(\Omega),$$

(1)

where Ω' and Ω are the angular frequency detunings from the center frequencies of the fundamental and the SH waves, respectively, and A_1 and A_2 are the Fourier transforms of the field envelope functions of the fundamental and the SH, respectively. For a uniform nonlinear crystal of length L, $D(\Omega)$ is given as

$$D(\Omega) = \Gamma L \operatorname{sinc}(\Omega \alpha L/2),$$

(2)

where Γ is the nonlinear coupling coefficient and $\alpha = 1/v_{g1} - 1/v_{g2}$ arises from the GVM between the fundamental pulse (with group velocity v_{g1}) and the SH pulse (with v_{g2}). The GVM leads to a temporal broadening of the SH pulse. In the spectral domain this effect, described by $D(\Omega)$, limits the possible SHG frequency bandwidth to ~$0.88/(|\alpha|L)$.[6] It can lead to strong narrowing of the SH spectrum in thick crystals with large GVM (i.e., $L \gg \tau/|\alpha|$, where τ is the pulse width of an unchirped pump pulse). These effects are usually considered undesirable, and therefore most femtosecond SHG experiments are performed with thin crystals ($L \ll \tau/|\alpha|$).

Here we consider the situation in which $|\alpha|L \gg \tau$, such that $D(\Omega)$ approaches a δ function. Then the output SHG power is given as

$$\int_{-\infty}^\infty |A_2(\Omega)|^2 \mathrm{d}\Omega = 2\pi\Gamma^2 L$$

$$\times \left| \int_0^\infty A_1(\Omega')A_1(-\Omega')\mathrm{d}\Omega' \right|^2 /|\alpha|, \quad (3)$$

i.e., it is proportional to the self-convolution of the complex pump spectrum. It should be noted that the transition probability for two-photon absorption in an atomic medium has the same functional form as Eq. (3), assuming no intermediate resonance; this fact has formed the basis for experiments on coherent quantum control of two-photon transitions by use of pulses shaped by sinusoidal spectral phase modulation.[7] Therefore our experiments can be regarded as a nonlinear optical but completely classical analog for the weak-field quantum-control experiments observed in the research reported in Ref. 7. However, broadband two-photon processes, such as SHG in thin crystals and two-photon absorption into a continuum,[12] are fundamentally different; they depend only on the temporal intensity profile.

Equation (3) also shows that the SH power scales linearly with L. Therefore, increasing L can lead to highly efficient SHG of femtosecond pulses, even under conditions of large GVM.[13]

Equation (3) can be further simplified if the input spectrum is flat-topped and unchirped. Under such

conditions spectral coding can be realized with the scheme shown in Fig. 1. The fundamental spectrum is divided into $2N$ channels with equal frequency widths, each of which is given a phase shift Φ_i $(i = -N, \ldots, -1, 1, \ldots, N)$. Thus the SHG signal is given by

$$P_{\text{SHG}} \propto \left| \sum_{i=1}^{N} \exp(j\Phi_i)\exp(j\Phi_{-i}) \right|^2 . \qquad (4)$$

If we divide the spectrum in half and call the phase patterns on the low-frequency and high-frequency sides of the spectrum C_1 and C_2, respectively, the SHG signal is proportional to the square of the magnitude of the correlation between code words $C_1 = \{\Phi_{-1} \ldots \Phi_{-N}\}$ and $C_2 = \{\Phi_1 \ldots \Phi_N\}$. Therefore one can control the SHG yield by changing the correlation properties of the applied spectral phase functions.

Our experimental setup consisted of a passively mode-locked fiber laser, a fiber-pigtailed femtosecond pulse shaper[9] with a 128-pixel programmable amplitude–phase liquid-crystal modulator array,[5] a 20-mm-long periodically poled lithium niobate crystal placed in a temperature-controlled oven, and a spectrometer. Periodically poled lithium niobate is ideal for our application because of its high nonlinearity, large GVM parameter (0.3 ps/mm), and noncritical phase matching that avoids spatial walk-off.[11] The output pulses from the pulse shaper had an energy of ~2 pJ at a 40-MHz repetition rate. The liquid-crystal modulator was programmed to equalize the pulses' spectral amplitude and compensate for any residual spectral phase chirp from the fiber system[14] in addition to applying the spectral phase codes. The resultant spectrum was approximately flat, with a width of ~18 nm centered at ~1559.5 nm. The SH phase-matching wavelength was temperature tuned to 779.75 nm. The light was loosely focused into the crystal, with a depth of focus longer than the crystal. The SHG output was measured with the spectrometer. For uncoded pulses the SHG generated from the equalized, unchirped pump input had the expected sinc^2 shape with a bandwidth of ~0.3 nm.

We studied two different families of binary codes (M-sequence and Hadamard codes) known from communications theory.[15,16] To convert the binary codes into phase codes we set Φ_i to π when the corresponding code bit was -1 and to 0 when the bit was 1. In our experiments C_1 was set to a fixed code word in a code set while C_2 was varied among all the words in that set. The spectral coding process spread the unchirped femtosecond input pulse into lower-intensity waveforms ~10 ps long and with complicated substructures. Shown in Fig. 2 are experimental intensity cross-correlation measurements as well as theoretical intensity profiles for coded pulses that use Hadamard codes. Data are shown for a fixed C_1 and two different choices of C_2. In each case we can see a close match between the calculated waveforms and the data. Even though the intensity profiles look quite similar, the correlation properties of the concatenated codes are quite different. One of them [Fig. 2(a)] consists of highly correlated code words (in this case

$C_1 = C_2$), which should yield the same SH intensity at the center frequency ($\Omega = 0$) as in the uncoded case. The other [Fig. 2(b)] consists of two uncorrelated code words, which result in weak SH at $\Omega = 0$.

As a first case, we present results for a set of eight length-8 Hadamard codes,[16] which are orthogonal in their bipolar form and therefore should strongly suppress the SHG signal at $\Omega = 0$, ideally to zero. C_1 was set to be the second code word $[-1\,1\,-1\,1\,-1\,-1\,1]$ in the code set. Figure 3(a) shows the measured SHG spectrum for different choices of C_2. For coded pulses, in the situation with correlated codes ($C_1 = C_2$, as in curve II), strong SHG was observed, and its peak was comparable with that of the uncoded case (curve I), even though the input pulse had been dispersed significantly in time. The SHG bandwidth was then dominated by the correlation function of the code and was narrower than in curve I. The minor difference
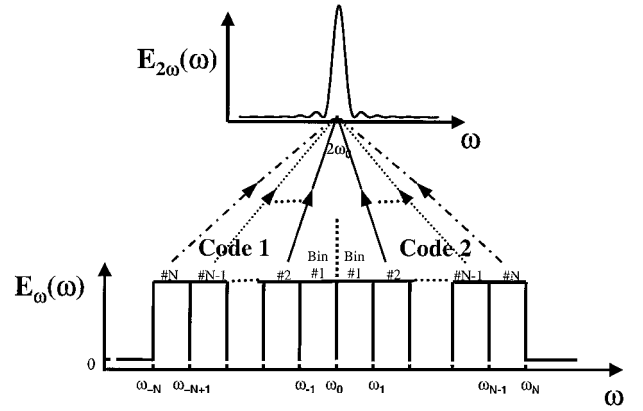

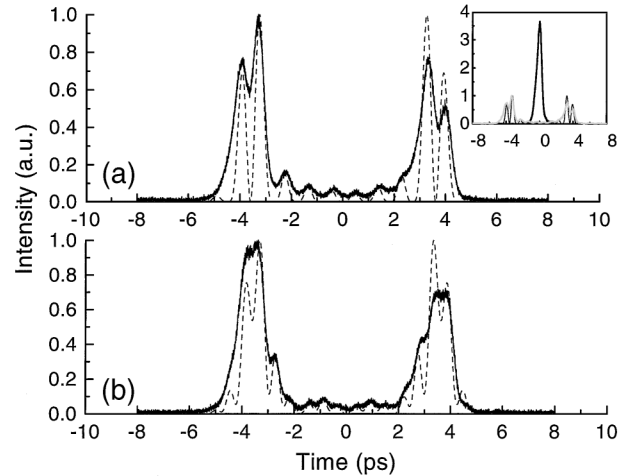Fig. 1. Schematic of optical phase coding for the control of SHG by ultrashort pulses.

Fig. 2. Theoretical intensity profiles and experimental intensity cross-correlation data of spectrally coded pulses. (a) Phase code $\{1\,-1\,1\,-1\,1\,-1\,1\,-1 : -1\,1\,-1\,1\,-1\,-1\,1\,1\}$ was used, and it consisted of C_1 and C_2 with the bits in C_1 arranged from right to left and the bits in C_2 arranged from left to right (as shown in Fig. 1). Here C_1 and C_2 were the same: $\{-1\,1\,-1\,1\,-1\,-1\,1\,1\}$, the second code word in the Hadamard matrix (see Ref. 16). Inset, uncoded pulse versus coded pulse. (b) Phase code $\{1\,-1\,1\,-1\,1\,-1\,1\,-1 : -1\,1\,-1\,1\,-1\,1\,-1\,1\}$ was used, with C_1 kept the same and C_2 being the sixth word: $\{-1\,1\,-1\,1\,1\,1\,-1\,1\}$.
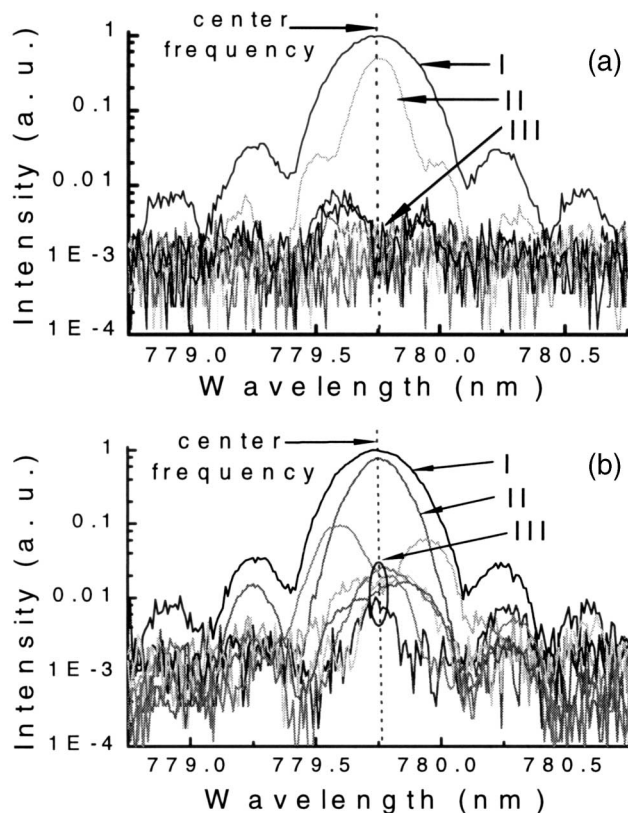
Fig. 3. SHG spectra measured in different spectral phase-coding situations: I, no phase code was applied to the pump pulses; II, phase codes C_1 and C_2 are the same; III, data for fixed C_1, for all choices of $C_2 \neq C_1$. (a) C_1 and C_2 chosen from length-8 Hadamard codes; (b) C_1 and C_2 chosen from length-7 M-sequence codes, given by all the cyclic permutations of $\{-1-1111-11\}$.

in the peak intensities of curves I and II may arise from small imperfections in the coding process. The seven traces that correspond to $C_1 \neq C_2$ show weak SHG. Compared with that in the uncoded-pulse case, the SHG at the center frequency was suppressed by at least 600 for all seven unmatched codes and by more than 1500 in some traces (limited by the noise floor of our measurements). We note that, if a longer nonlinear crystal is used with a SHG bandwidth narrower than the correlation function of the codes, the total SHG yield can be controlled even without a spectrometer, with the contrast ratio given by relation (4).

As a second case, we used a family of seven length-7 M-sequence codes, which have a ratio of autocorrelation to cross correlation that is equal to the code length. Thus there should be a contrast ratio of 49 between matched and unmatched codes. For each of the six cases when $C_1 \neq C_2$, the SHG was suppressed by a factor from ~ 40 to ~ 100 at the center wavelength, in reasonable agreement with theory, although the spectra have vastly different shapes at other wavelengths. Comparing the M-sequence and the Hadamard code results, we note that because of the pseudorandom nature of the M-sequences the intensity profiles of pulses coded with M-sequences[9] are much more uniformly spread in time, with lower

peak intensities than for Hadamard codes. Therefore, for broadband SHG or two-photon absorption into a continuum, M-sequence coding would give the greater suppression (even so, the contrast between uncoded and coded waveforms is proportional only to the code length, which gives weaker suppression than what is observed here), in contrast to our results here. These observations underscore the strong differences between control of narrow-band SHG, which makes use of constructive and destructive interference between the pathways indicated in Fig. 1, and broadband SHG, which is sensitive only to the optical intensity.

In summary, we have demonstrated high-contrast, accurate control in femtosecond SHG by optical phase coding. Our results demonstrate the possibility of realizing a spectral phase correlator that could play an important role in ultrashort-pulse code-division multiple-access networking schemes.[8-10] The results demonstrated here, for which entire families of codes taken from communications theory were used, should also be applicable to coherent control of two-photon transitions in gases,[7] which has a similar mathematical basis. For practical applications, however, the SHG scheme has advantages in terms of sensitivity and ease of implementation.

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