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Filling a spectral hole via self-phase modulation

A. Präkelt, M. Wollenhaupt, C. Sarpe-Tudoran, A. Assion, and T. Baumert[a)]
University of Kassel, Institute of Physics and Center for Interdisciplinary Nanostructure Science and Technology (CINSaT), Heinrich-Plett-Str. 40, D-34132 Kassel, Germany

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The effect of spectral amplitude modulation on self-phase modulation is studied. To that end we remove a small interval of frequency components from the broad spectrum of a femtosecond laser pulse. We investigate the regeneration of these missing frequency components via self-phase modulation. A water jet serves as a transparent sample. A physical model is given which explains the observation that the removed frequency components are not only replenished by self-phase modulation but can even overshoot their adjacent frequencies in power spectral density. In addition, we suggest possible applications in the field of nonlinear microscopy. *© 2005 American Institute of Physics*. [DOI: 10.1063/1.2056589]

Self-phase modulation (SPM) is an important ultrafast nonlinear optical process that plays a dominant role in continuum generation. It has been a field of intense research since its discovery in the 1960s.[1,2] Continuum generation has found a large variety of applications. Among these are, for example, well established techniques such as pulse compression and transient absorption spectroscopy as well as emerging applications like frequency metrology,[3] optical coherence tomography[4] and femtosecond remote sensing.[5] Recently, pulse shaping techniques were applied to control the continuum generation in microstructured fibers.[6] Besides SPM, a variety of other effects such as self-focusing, self-steepening, stimulated Raman scattering and four-wave mixing are studied in the field of continuum generation.[1,2]

SPM creates new frequency components during the nonlinear interaction of an ultrashort laser pulse with a transparent sample. In our contribution, we remove a narrow band of frequency components from the spectrum of a femtosecond laser pulse employing spectral pulse shaping techniques.[7] The missing frequency components are regenerated by SPM in a thin water jet serving as a transparent sample. Water was chosen as a convenient nonlinear medium and as a first approximation to possible biological applications. Observing the power spectral density (PSD) as a function of the laser intensity, we show that the removed frequencies are not only replenished but can even overshoot their adjacent frequencies. We reproduce this effect in a simple simulation taking only SPM into account and provide a physical model of the effect.

The experimental setup is depicted in Fig. 1. A Ti:sapphire amplifier provided femtosecond laser pulses of 30 fs full width at half maximum (FWHM) duration (measured at the location of the sample) with a repetition rate of 1 kHz. The femtosecond laser pulses were amplitude modulated with the help of our home-built pulse shaper.[8] For this experiment, we replaced the liquid-crystal spatial light modulator by a 0.2-mm-thick wire allowing us to eliminate a narrow spectral band of 2 nm. Because the pulse energy should be reduced only by a small amount and the experiment should be close to the physical model presented later on, the diameter of the wire was chosen reasonably narrow. It approximately matches the spectral spot size of the beam waist in the Fourier plane of the pulse shaper (1 nm) in accordance with Ref. 9. Experimentally, we found that the best performance is achieved by placing the wire at 788 nm outside the spectral region with highest PSD (λ_0). This observation is confirmed by the theoretical findings (see below). The amplitude modulated femtosecond laser beam was focused by a 50 mm lens into a 100-μm-thick distilled water jet. We measured the thickness of the jet via spectral interference of the reflected light. The beam waist radius of 15 μm was obtained by a cutting knife method. The transmitted beam was collected by a $10\times/0.28$ NA infinity corrected objective at a distance of 3 cm from the jet and focused by a 200 mm lens into a fiber-coupled spectrometer with a charge coupled device detector. The intensity of the laser beam was adjusted by a motor controlled neutral density attenuator in front of the 50 mm lens and measured by a power meter. We varied the pulse energy from 0.5 to 5 μJ.

Reasonable accordance with our experimental results is achieved in our simulation. First, in this simulation, a 2 nm band of frequencies outside the central frequency is removed from the spectrum of a short Gaussian laser pulse of 30 fs FWHM duration. The spectral electric field is Fourier transformed into the time domain. In Fig. 2(a) we present the normalized temporal electric field envelope. The modulated pulse consists of a slightly attenuated original pulse at time zero and a sinc-type wing structure of preceding and following subpulses with a periodicity in the order of 1 ps. Since the sample thickness of 100 μm is smaller than the Rayleigh range (about 1.2 mm) and the self-focusing length (about 2.3 mm),[10] we consider SPM in the short sample approximation[1,11]

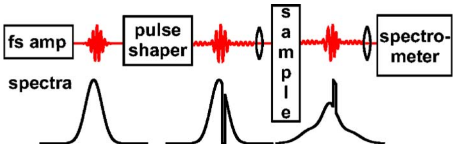

FIG. 1. Experimental setup: The femtosecond laser pulses from a Ti:sapphire amplifier (fs amp) are spectrally amplitude modulated using a pulse shaper. A lens focuses the beam into a water jet which serves as the transparent sample. The transmitted self-phase modulated beam is collected by an objective and focused into a spectrometer. In the lower row, schematic spectra of the pulses are depicted.

[a)] Author to whom correspondence should be addressed; electronic mail: baumert@physik.uni-kassel.de

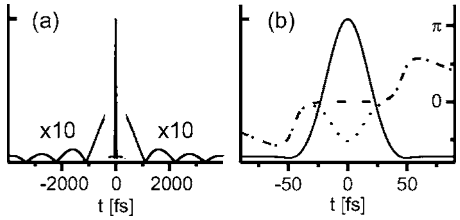

FIG. 2. Normalized temporal electric field envelopes of a Gaussian pulse with removed frequencies. (a) Shows the short central part and the ten times magnified wing structure without self-phase modulation, (b) focuses on the central part. In this part, the temporal phase (plotted without carrier) is flat without self-phase modulation (dashed line) and bell shaped with self-phase modulation (dotted line). The intensity corresponds to Fig. 3(d).

$$E_{\mathrm{SPM}}(t) = \mathcal{E}(t)\cos[\chi(t) + \chi_{\mathrm{SPM}}(t)] \tag{1}$$

with the self-phase modulated temporal electric field $E_{\mathrm{SPM}}(t)$, the temporal envelope $\mathcal{E}(t)$ and phase $\chi(t)$ of the electric field with the frequencies removed. The additional temporal phase generated due to SPM is given by

$$\chi_{\mathrm{SPM}}(t) = -\frac{2\pi}{\lambda_0}\frac{n_2}{n_0} d I(t), \tag{2}$$

where $n_0 = 1.33$ and $n_2 = 5.7 \cdot 10^{-20}$ m^2/W are the linear and nonlinear refractive indexes for distilled water.[12] The term $d = 100$ μm is the thickness of the sample and $I(t)$ is the temporal intensity of the electric field with removed frequencies. Figure 2(b) depicts the normalized envelope and the phase of a self-phase modulated pulse. In our case, SPM does not change the shape of the temporal electric field envelope. The phase of the pulse without SPM is shown as a dashed line in Fig. 2(b) and is flat around time zero. According to Eq. (1), SPM introduces an additional phase which is proportional to the square of the envelope and, thus, increases with intensity. Therefore, due to SPM, the phase around time zero changes from flat to bell shaped while the phase at larger times changes only slightly [see dashed and dotted line in Fig. 2(b)]. Fourier transformation of the self-phase modulated temporal electric field yields the spectral electric field and the PSD.

In Fig. 3, we show our experimental and corresponding simulated normalized spectra. In addition to the PSD, the spectral phase (dashed line) is shown in selected cases of the simulations. The intensities of the pulses are given in units of a reference intensity. Due to the specific shape of the temporal electric field (see Fig. 2) the values for the intensity are dependent on details of the averaging procedure. For instance, the experimental I_0^{exp} and theoeretical reference intensities I_0^{th} are: (1) $I_0^{\mathrm{exp}} = 1.5 \cdot 10^{13}$ W/cm^2 and $I_0^{\mathrm{th}} = 5 \cdot 10^{12}$ W/cm^2 for the original unmodulated pulse with all frequencies, (2) about 5% less for the central temporal part of the pulse when the frequencies are removed [see Fig. 2(b)], (3) $I_0^{\mathrm{exp}} = 9 \cdot 10^{11}$ W/cm^2 and $I_0^{\mathrm{th}} = 3 \cdot 10^{11}$ W/cm^2 for the whole pulse with removed frequencies, employing a statistical definition of the pulse duration.[11] The intensities of the unmodulated pulse might be close to the tolerable limit of biological samples. The spectra at low intensity without SPM are displayed in Fig. 3(a). In the experiment, the water jet was switched off. The missing frequency components at 788 nm result in a "spectral hole." The spectral phase is flat. In Fig. 3(b), the jet was switched on. Due to SPM the removed frequencies are slightly regenerated. With increasing intensity, the removed frequencies are gradually regenerated [see Fig. 3(c)]. At the reference intensities I_0^{exp} and I_0^{th}, respectively, the spectral hole has been replenished as depicted in Fig. 3(d). SPM results in a bell-shaped spectral phase while the removal of a spectral band causes a phase jump at the removed frequencies. The general shape of the spectral phase does slowly change with increasing intensity. At even higher intensities, the removed frequencies overshoot their adjacent frequencies creating a structure similar to a "spectral hill" [see Fig. 3(e)]. The additional structures in the experimental spectra of higher intensities are very sensitive to higher order chirp contributions as investigated in independent experiments.

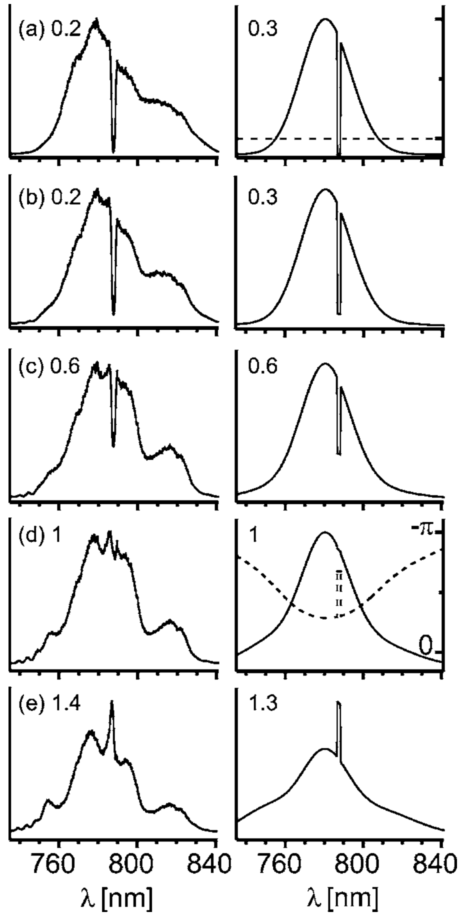

FIG. 3. Experimental (left) and simulated (right column) normalized power spectral densities and simulated spectral phases (dashed lines). The intensities are shown in the upper left corners (in units of I_0^{exp} and I_0^{th}). In case (a), the water jet was switched off (no self-phase modulation) while it was turned on in all other cases.

We now discuss the effect of spectral overshoot in a simplified model based upon Fourier decomposition arguments. In Fig. 4(a), the broad spectrum and the short temporal electric field of a 5 fs model pulse are depicted. The temporal electric field is decomposed into the modes of the pulse which are also shown. In Fig. 4(b), certain frequency components are removed from the spectrum. In the time domain, this removal can be interpreted as destructive interference of the corresponding modes with sinc-type quasi-continuous waves (QCWs) whose phase is shifted by π. In the experiment, the QCWs would have a limited temporal duration due to the finite spectral resolution of a pulse shaper.[9] The ultrashort laser pulse is superimposed by QCWs. In the time domain, the effect of SPM is described

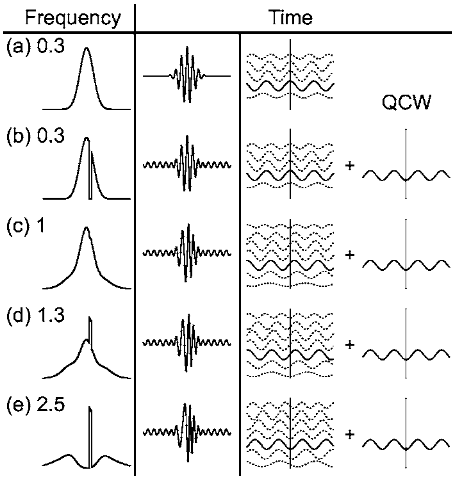

FIG. 4. Schematic physical model of spectral hole filling via self-phase modulation using 5 fs pulses for visualization. In the first column the spectra of the electric fields are depicted. The second column shows the temporal electric fields. In the third column the Fourier decompositions of the pulses into their modes are sketched. The time zeroes are marked by vertical solid lines. The removal of a certain frequency component (mode with solid black line) is interpreted as the interference with a quasi-continuous wave (QCW) [see cases (b)–(e)]. In (c)–(e) the effect of self-phase modulation is shown for increasing intensity (given in units of I_0^{th}).

by the alternation of the temporal phase of the ultrashort laser pulse [see Eq. (1)]. However, a physical picture of the hole filling process is given in the frequency domain. Because SPM generates a linear chirp (to lowest order approximation) the spectral phase of the nonamplitude modulated pulse with all frequencies is a bell-shaped function. For the spectrally amplitude modulated pulse with removed frequencies, the jump in the spectral phase at the removed frequencies results from the complex summation of the spectral amplitudes of the self-phase modulated pulse and the π-shifted QCWs [see Fig. 3(d)]. Since the QCWs have a very low intensity in comparison to the pulse the phases of the QCWs are not influenced by SPM. Because SPM changes the spectral phase more efficiently outside the central frequency [see Fig. 3(d)] the relative phase between the modes of the pulse and the QCWs changes more rapidly at that spectral location. With increasing intensity, this interference between the modes and the QCWs changes from completely destructive to constructive. In the frequency domain, this means that the removed frequency components are regenerated. This is illustrated in Fig. 4(c) for the case of replenished frequency components. With further increasing pulse intensity, the constructive interference between the modes and the QCWs becomes more pronounced so that a spectral overshoot is generated [Fig. 4(d)]. In addition, the case of completely constructive interference where the modes and the QCWs are in phase is shown in Fig. 4(e) (not measured). Here, the initially removed frequencies strongly overshoot their adjacent frequencies in PSD. For even higher intensity, the interference becomes once again destructive. A related mechanism was recently reported to explain atomic transitions with missing frequencies in a THz spectrum.[13]

We plan to investigate to what extent the effects of spectral replenishment and overshoot can be used as a new contrast mechanism in nonlinear microscopy of transparent samples for the visualization of different n_2 regions. Within this context pulse shaping techniques have been used to enhance two-photon microscopy[14,15] and coherent anti-Stokes Raman spectroscopy[16] which has also been applied as a microscopy technique.[17] Imaging index matched transparent samples has been demonstrated with the help of third harmonic generation microscopy.[18] Our approach would be based on SPM in combination with spectral amplitude modulation. The contrast could be obtained from the ratio between the missing frequencies and the adjacent frequencies. For example, this ratio changes from zero to nearly 2 with increasing intensity in Fig. 3. This contrast scheme could be particularly suitable to the investigation of biological tissues since both excitation and detection are in the transparency window of most biological tissues.

In conclusion, we have investigated the effect of spectral amplitude modulation on self-phase modulation. Frequency components which are removed from the spectrum of an ultrashort laser pulse by spectral amplitude modulation are regenerated via self-phase modulation. With increasing intensity, the removed frequencies are replenished and even overshoot their adjacent frequencies in power spectral density. This effect was discussed in the view of a physical model based on the Fourier decomposition of an ultrashort laser pulse into its modes and the interference with additional sinc-type quasi-continuous waves. Possibly, our experiment can be extended to provide contrast in nonlinear microscopy.

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