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Comb-like supercontinuum generation in bulk media

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We report the generation of sequences of phase-coherent white-light pulses in bulk media. By passing the pulses from an amplified femtosecond laser through an etalon cavity, we produce an equally spaced time sequence of phase-locked pulses that serve as the pump for the generation of supercontinuum. The mutual coherence among the white-light pulses is probed by studying their spectral interference patterns for varying pump energy levels. © *2004 American Institute of Physics*.
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The ability to generate wide-bandwidth optical frequency combs by using femtosecond mode-locked lasers has recently provided a revolutionary advance in the progress of metrology and optical frequency synthesis.[1] The frequency spectrum of the regular pulse train emitted by a mode-locked laser consists of a comb of modes whose spacing is simply determined by the repetition rate of the laser and extends over a frequency range which scales inversely with the pulse duration. Femtosecond frequency combs are now replacing old-style frequency chains wherever a precise measurement of an optical frequency is required because, with the realization of combs so wide as to extend over more than one optical octave, this technique now constitutes a solid self-referencing method, allowing the measurement of absolute optical frequencies with extreme accuracy in a single step from the microwave frequency standard. Since such a wide bandwidth would correspond to a transform-limited single-cycle pulse which has not been achieved yet at optical frequencies, all recent results made use of an external nonlinear process to broaden the spectrum.[2–4] The simplest way to achieve such an extreme spectral broadening is by the process of white-light supercontinuum (SC) generation: since the first observations in the late 1960s,[5] this phenomenon, occurring when ultrashort and powerful laser pulses propagate in a transparent medium, has been demonstrated in a variety of materials, including solids, liquids,[6] and gases.[7,8] SC generation is the result of a delicate balance among several nonlinear optical effects (self-phase-modulation[5,6] and self-focusing[7,9] being the main phenomena involved), and it might seem highly dependent on the exact initial conditions of interaction with the medium. In particular, one is led to expect that even small perturbations, such as intensity fluctuations in the pump pulses or small inhomogeneities in the materials, may strongly affect the phase and intensity of the generated SC pulses. Such effects are not expected to be so dramatic in the case of SC generation in microstructured fibers, where the pulse intensities are normally kept relatively low and the nonlinear broadening effect is accumulated over a long guided propagation distance. On the other hand, the generation of SC in bulk media requires the substantially higher intensities available with amplified laser pulses and the process takes place under far less controlled conditions. The preservation of the phase coherence in the process of white-light generation in bulk media has however been recently demonstrated in several different configurations: in the case of two spatially separated sources,[10–12] for a pair of collinear SC pulses,[13] and for a linear array of SC sources.[14] In this work we demonstrate a small-scale version of a femtosecond frequency comb by generating a "short" train of equally spaced and phase-locked SC pulses in a bulk material and studying their combined spectrum as a function of the pump pulse power.

The pump laser for the experiment is an amplified Ti:sapphire laser system delivering 0.8 mJ, 30 fs pulses, centered at 780 nm and at repetition rate of 1 kHz. The output beam is apertured by an iris and directed toward an etalon cavity (made of two facing fused silica mirrors, air-spaced by a distance $d=25$ μm). Because of the high reflectivity ($R=0.9$) of the etalon surfaces, only about 10% of the incident radiation enters the cavity and only another 10% of it exits the etalon after a backreflection on the internal surface. Since the temporal coherence of the laser pulses is much shorter than the cavity round-trip time $t_r=2d/c\approx 167$ fs, the leading edge (having been reflected twice by the cavity mirrors) does not interfere with the trailing edge of the pulse itself, and the output time structure after multiple reflections between the cavity mirrors is thus simply given by a sequence of pulses centered at $t_j=jt_r$ and with decreasing energies E_j, such that

$$E_j=E_0R^{2(j-1)}(1-R)^2, \qquad (1)$$

where E_0 is the initial laser pulse energy. The ratio between the energies of successive pulses exiting the cavity is thus given by $E_{j+1}/E_j=R^2\approx 0.81$. A lens is used to focus the pulse sequence into a 1-mm-thick CaF_2 plate, while another lens focuses the generated white light onto the entrance slit of a spectrometer, based on a 2048 pixels photodiode array, with a spectral coverage extending from 520 to about 1180 nm and a spectral resolution of 1.5 nm (see Fig. 1). The CaF_2 plate is continuously moved in a plane perpendicular to the propagation direction by the combination of two sinusoidal linear oscillations at incommensurable frequencies. The resulting speed (of the order of 10 cm/s), combined with a beam waist diameter of 30–40 μm, allowed different pulse trains (spaced by 1 ms) to always propagate in a "fresh" medium in order to avoid the possible damage related to the high peak power.

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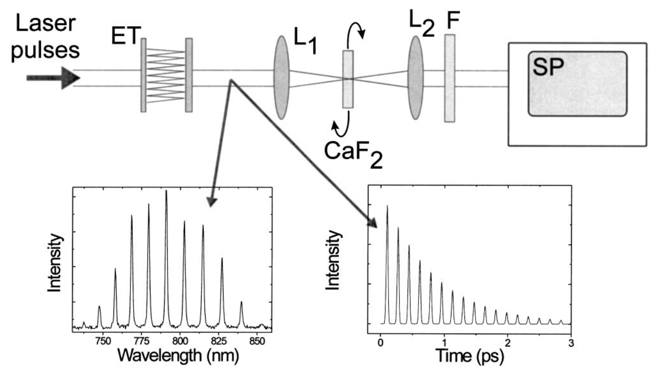

FIG. 1. Experimental apparatus ET: etalon cavity; L_1 and L_2: lenses; F: filter; SP: spectrometer. Also shown are an experimental spectrum of the laser after passing through the etalon (left graph) and the calculated intensity profile of the output laser pulse sequence (right graph).

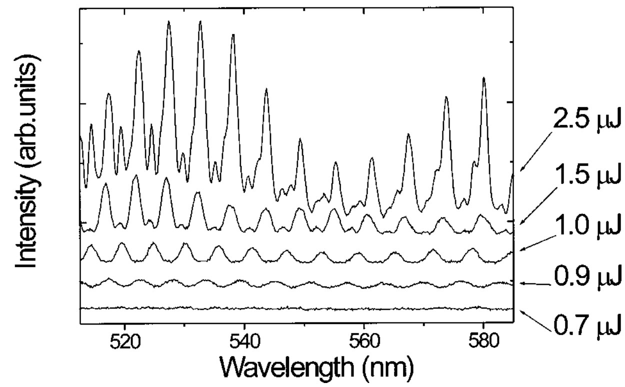

FIG. 2. Plots of the white-light spectra at different values of the energy E_1 of the first pulse in the sequence. Traces have been vertically displaced for improved readability.

At low incident powers (<50 mW as measured before the etalon), the spectrum of the pulse sequence at the exit of the cavity and after the interaction with the medium is simply given by the product of the Gaussian laser spectrum multiplied by the Airy formula for the normal incidence transmission through the etalon:

$$I_T(\omega) = I_0 \exp\left(-\frac{(\omega-\omega_0)^2}{\Delta\omega^2}\right)\frac{1}{1+F\sin^2\left(\frac{d\omega}{c}\right)}, \qquad (2)$$

where ω_0 is the laser central frequency, $\Delta\omega$ is the $1/e$ laser spectral half-width, and $F=4R/(1-R^2)$ is the Finesse of the cavity (≈ 19 in our case), also expressed as the ratio between the cavity free-spectral-range ($\Delta\lambda=\lambda^2/2d$, corresponding to about 12 nm around the laser central wavelength) and the width of the etalon transmission peaks ($\delta\lambda \approx 0.64$ nm). In such conditions, the energy $E_1 \approx 0.5$ μJ of the first pulse exiting the etalon cavity is still just below the threshold for SC generation, and no spectral broadening is observed after the interaction with the material. If one assumes that SC is initiated by the spatial collapse of the beam due to self-focusing and by the formation of a filament, then one can estimate a critical pulse energy E_c for the sudden onset of spectral broadening of the order of 0.6 μJ. When the laser power is increased so that $E_2 < E_c < E_1$, it is possible to satisfy the conditions of spectral super-broadening for the first pulse of the sequence only, while leaving the successive pulses almost unaffected. Things start to get more interesting when the input pulse energy is increased to such a level ($E_2 > E_c$) that also the second pulse of the sequence becomes able to generate white light. If the two delayed SC pulses maintain a memory of the phase of the generating laser pulses (i.e., if the phase of the pump pulses is transferred to the SC pulses without significant distortions or, at least, with identical distortions in the two), then they are expected to interfere when observed in a spectrally resolved fashion. The spectrum of the resulting light should thus be sinusoidally modulated, with an initially low fringe visibility due to the intensity imbalance of the two interfering SC pulses; the visibility will then grow toward a maximum close to unity as the two SC pulses tend to reach a common saturation intensity. Note that, thus far, the situation is similar to the one already presented in Ref. 13, where the two collinear and time-delayed SC pulses were produced by means of a Michelson interferometer. In the present experiment, however, the number of collinear and equally time-delayed SC pulses can be increased and simply controlled by varying the energy of the pump laser pulses. When such energy is further increased, successive pulses from the sequence exiting the etalon cavity start generating supercontinuum and may contribute to the spectral interference pattern. If the mutual phase-lock is preserved for a train of N white light pulses, then one should expect a complex multiple-beam interference pattern, essentially composed of sharp spectral maxima, whose width decreases like $1/N$, spaced of $\Delta\lambda$, with $N-2$ smaller (by a factor of about N^{-2}) peaks interposed. Figure 2 shows the spectra of the white light SC around 550 nm as observed for increasing values of the energy E_1 of the first pulse exiting from the cavity. The passage from a single SC pulse with a relatively flat spectrum for a pulse energy of about 0.7 μJ, to a pair of pulses showing a clear sinusoidal spectral modulation at 0.9 μJ, to more complicated situations with the presence of secondary peaks and narrower main maxima, is evident. The limited resolution of the spectrometer prevents us from observing the finer structures involved in the multiple-pulse interference, but the onset of SC generation from three successive pulses is nonetheless clear for $E_1=1$ and 1.5 μJ, while more than four phase-locked SC pulses are evidently responsible for the spectral interferences present at $E_1=2.5$ μJ. Note that a variation in the number of interfering sources can be obtained not only by changing the initial pulse energy, but also by looking at different spectral regions at a fixed energy value. The broadening process indeed proceeds from the laser wavelength outwards, so that for a given initial energy, the number of pulses contributing to SC generation decreases by moving away from the laser spectrum. The spectrum may thus start flat in the extreme wings of the distribution and then develop sinusoidal modulations which finally get sharper while approaching the laser wavelength. Such effect is clearly visible in the measured spectra covering wider frequency intervals as reported in Fig. 3. Long integration times can be used to record the spectral distributions without substantially degrading the visibility of the interference patterns. The high robustness and stability of the observed spectral features is a proof of the role of self-stabilizing processes taking place during the formation of filaments as a fundamental ingredient for reliable SC generation. It is nonetheless interesting to note that the length of the train of interfering SC pulses cannot be extended indefinitely by increasing the laser pulse energy. Indeed, when the jth pulse of the sequence starts generating white light, the first

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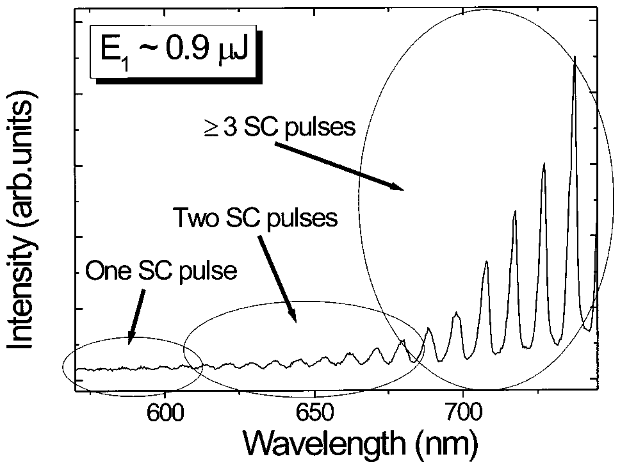

FIG. 3. White-light spectrum showing the increase in the number of interfering SC sources while getting closer to the laser wavelength.

pulse must have already reached an energy level which is at least $R^{2(1-j)}$ times higher than the threshold E_c, and may thus start to cause unwanted additional effects, such as multiple filamentation and material damages. When such conditions are reached, the SC generation process is no longer as stable and smooth as necessary for an accurate phase-lock among the different pulses and the resulting spectra lose their regularity. On the other hand, a change in the temporal spacing among the different pulses in a train (obtained for example by using a longer cavity length) is not expected to cause significant differences in the results, apart from a change in the spacing of the spectral peaks that may prevent observation in the case of limited resolution of the spectrometer. A rather accurate simulation of the experimental results has been obtained by using a simple model where a quadratic phase modulation with a nonlinear coefficient showing a characteristic threshold behavior as a function of the incoming pulse power is introduced. A more detailed analysis of the observed spectra will be the subject of a forthcoming paper.

In conclusion, we have demonstrated the generation of a small-scale version of a femtosecond frequency-comb operating at high peak intensities in bulk materials. The existence and the excellent stability and visibility of the resulting multiple-pulse spectral interferences are a proof of a stable phase-lock among the different SC pulses in the sequence.

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[1] S. T. Cundiff, J. Ye and J. L. Hall, Rev. Sci. Instrum. **72**, 3749 (2001).
[2] J. Reichert, M. Niering, R. Holzwarth, M. Weitz, Th. Udem, and T. W. Hänsch, Phys. Rev. Lett. **84**, 3232 (2000).
[3] D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Wideler, J. L. Hall, and S. T. Cundiff, Science **288**, 635 (2000).
[4] R. Holzwarth, Th. Udem, and T. W. Hänsch, Phys. Rev. Lett. **85**, 2264 (2000).
[5] R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. **24**, 584 (1970).
[6] R. L. Fork, C. V. Shank, C. Hirlimann, and R. Yen, Opt. Lett. **8**, 1 (1983).
[7] P. B. Corkum, C. Rolland, and T. Srinivasan-Rao, Phys. Rev. Lett. **57**, 2268 (1986).
[8] F. A. Ilkov, L. Sh. Ilkova, and S. L. Chin, Opt. Lett. **18**, 681 (1993).
[9] W. L. Smith, P. Liu, and N. Bloembergen, Phys. Rev. A **15**, 2396 (1977).
[10] M. Bellini and T. W. Hänsch, Opt. Lett. **25**, 1049 (2000).
[11] W. Watanabe, Y. Masuda, H. Arimoto, and K. Itoh, Opt. Rev. **6**, 167 (1999); W. Watanabe and K. Itoh, Jpn. J. Appl. Phys., Part 1 **40**, 592 (2001).
[12] K. Cook, A. K. Kar, and R. A. Lamb, Appl. Phys. Lett. **83**, 3861 (2003).
[13] C. Corsi, A. Tortora, and M. Bellini, Appl. Phys. B: Lasers Opt. **77**, 285 (2003).
[14] C. Corsi, A. Tortora, and M. Bellini, Appl. Phys. B: Lasers Opt. **78**, 299 (2004).

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