Appl. Phys. B 77, 285–290 (2003)
DOI: 10.1007/s00340-003-1184-9

Applied Physics B
Lasers and Optics

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Mutual coherence of supercontinuum pulses collinearly generated in bulk media

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Received: 24 February 2003/Revised version: 15 April 2003
Published online: 6 June 2003 •

ABSTRACT We report the collinear generation of white light pulses in bulk media by phase-locked and time-delayed intense laser pulses. The mutual phase coherence of the pump pulses is fully transferred to the two supercontinua, which thus show a highly-modulated two-pulse spectrum extending from all of the visible down to the near-infrared. We have investigated the effects of changing the relative delay between the pulses as well as the role of imbalances in the pump energy, and we have found that, at least in a limited energy interval, the mutual phase coherence of the two white-light pulses is surprisingly robust.

PACS 42.65.Jx; 42.65.Ky; 42.65.Re

1 Introduction

Ultrashort and powerful laser pulses propagating in a transparent medium can undergo extreme spectral broadening, resulting in the generation of white-light pulses with frequencies ranging from the UV to the IR.

Since the first observations of supercontinuum generation in the late 1960's [1], this phenomenon has been demonstrated in a variety of materials, including solids, liquids [2], and gases [3, 4]. Its unique characteristics make the supercontinuum an ideal broadband ultrafast light source [5] for applications, and it is now routinely used for femtosecond time-resolved spectroscopy [6, 7], in optical pulse compression for the generation of ultrashort pulses [8, 9], or as a seed pulse for optical parametric amplifiers [10–12].

The results of some recent experiments, demonstrating that the phase coherence of the pump pulses is preserved in the process of supercontinuum generation [13], have triggered a wealth of new work, culminating in the realization of the so-called femtosecond frequency combs, which are now revolutionizing the fields of high-precision spectroscopy and metrology [14–17].

Regular trains of phase-locked ultrashort laser pulses, such as those emitted by a mode-locked laser, are a powerful tool for measuring optical frequencies with very high precision. Indeed, the comb of modes (spaced by the known pulse repetition rate) that constitutes the spectrum of such a laser can be used as a ruler in the spectral domain to measure large frequency differences with extremely high precision. However, in the initial version of this technique, the largest measurable frequency gap was determined by the bandwidth of the laser radiation and extremely short pulses, or some form of limited spectral broadening, had to be used in order to bridge large frequency gaps [18–20]. Moreover, the technique only gave access to relative measurements, relying on a nearby spectral reference to determine unknown frequencies.

A reference-free absolute frequency measurement across all of the visible spectrum calls for an octave-spanning frequency comb. In this case, once the repetition rate of the mode-locked laser is accurately measured in the radio-frequency domain, one can directly measure an optical frequency by bridging the gap between this frequency and its second harmonic [14], or absolutely calibrate the comb modes by measuring the beat note between the blue end and the second harmonic of the red end of the same comb [17]. Supercontinuum generation is a convenient way to simply broaden laser pulse spectra to more than an optical octave. A self-referencing femtosecond frequency comb can thus be realized if a train of collinear and, most importantly, phase-locked supercontinuum pulses is generated from the train of phase-locked pulses emitted by a mode-locked laser. Note that the coherent nature of the supercontinuum generation process is essential for ensuring that the comb spectral structure of the mode-locked laser is transferred to the white-light continuum.

The first experiments dealing with the mutual coherence of supercontinuum pulses were performed with amplified laser systems in bulk media, demonstrating that spatially separated white-light sources could exhibit high-visibility spatial interference fringes [13, 21, 22] and that the phase of the laser pump pulses was preserved in the generation process. It took some more time and the invention of special photonic-crystal-fibers [23], which allow the confined propagation of ultrashort pulses with low dispersion, in order to obtain spectral superbroadening with the low-energy pulses of a mode-locked laser.

Although the current success of frequency combs as the most precise devices for frequency and time measurements clearly demonstrates that trains of accurately phase-locked collinear white-light pulses are routinely generated in microstructured fibers, no indication has been given so far about

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the possibility of generating collinear phase-locked continua in a bulk medium. In such a case continuum generation takes place under quite different conditions and substantially higher intensities are at play. For spatially separated sources, the pump laser pulses do not interfere in the medium and each pulse generates white light independently of the other. In the collinear case, on the other hand, substantial optical interference can take place between the laser pulses for delays shorter than their coherence time and the high intensities reached at the interference maxima may damage the material and prevent stable and efficient generation.

Continuum generation indeed results from a complex interplay of self-phase-modulation [1, 2, 24], self-focusing [3, 25], and several other nonlinear optical effects. The collapse of the beam profile due to self-focusing is one of the main ingredients for the generation of the continuum when the power of the pump pulse reaches a certain threshold. The combined action of these highly nonlinear phenomena might lead to the conclusion that even small perturbations in the conditions of interaction with the medium can strongly affect the amplitude and phase properties of the pulses. Also in the case of delays longer than the temporal coherence of the pump pulses, the second pulse of the white-light pair has to be generated in a region that has already strongly interacted with the first one, and some degradation of the mutual coherence of the two supercontinua might be expected.

In this work we investigate the generation of time-delayed and phase-locked collinear supercontinuum pulses in a transparent medium and we find that, at least in a limited range of pump intensities around 10^{12} W/cm^2, their spectral profiles show surprisingly well contrasted fringes, a clear indication that the two secondary sources are highly mutually coherent.

2 Experimental

The experimental setup is sketched in Fig. 1 and was based on an amplified Ti:sapphire laser system (Femtolasers, Femtopower PRO) delivering 0.8-mJ 30-fs pulses centered around 780 nm at a repetition rate of 1 kHz.

The laser pulse intensity was reduced to the desired range by decreasing the power of the laser pumping the multipass amplifier, and the emitted pulses were directed towards a sta-

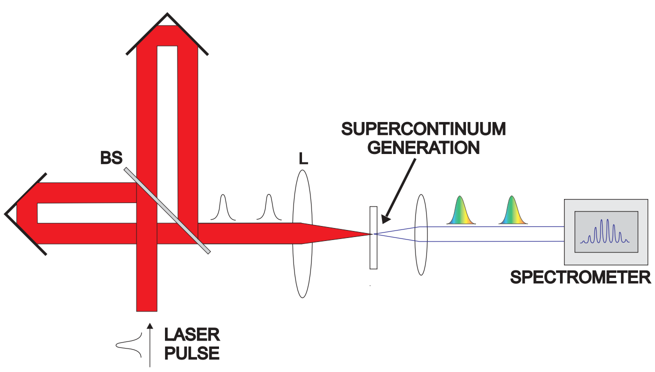

FIGURE 1 Experimental setup for the test of the phase-lock between collinear white-light continuum pulses. The IR pulses from the laser were split and recombined by a 50% beamsplitter (BS) and focused with a variable relative delay τ by the lens L in a glass plate. Frequency-domain interference fringes were detected by a spectrometer

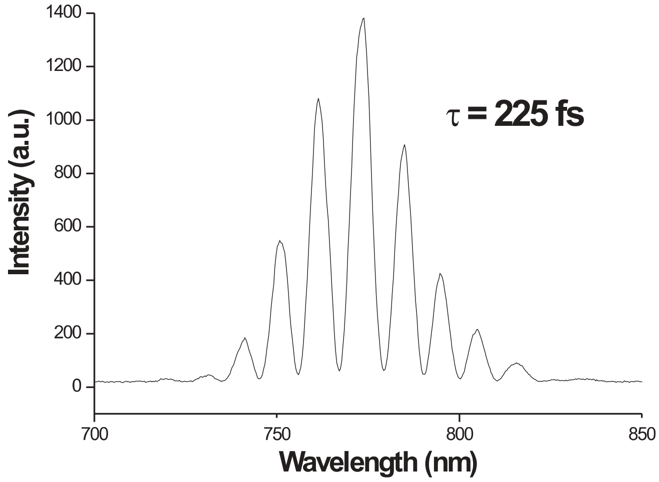

FIGURE 2 Interference fringes in the spectrum of the laser. The two laser pulses here were delayed by much more than their duration, so that they did not temporally overlap and interfere without a spectrally dispersive element

ble Michelson interferometer for the balanced generation of phase-locked and time-delayed pump pulses. The interferometer arm displacements were controlled by means of a high-precision computer-controlled dc motor and a piezoelectric (PZT) actuator. One of the mirrors of the interferometer was mounted on a PZT-controlled tilt stage for accurate alignment of the outgoing pulses; the laser pulses had to be exactly collinear in order to observe clear spectral interferences, and the optimization of the fringe contrast for the two-pulse laser spectrum was indeed used to maximize the beam overlap and collinearity (see Fig. 2). Spectra were observed after a re-collimating lens by means of a spectrometer, based on a 2048-pixel photodiode array, with a spectral coverage extending from 520 to about 1180 nm.

A 200-mm focal length lens was used to focus the laser pulses into the transparent materials; glass windows with thicknesses ranging between 0.5 and 5 mm were used for most of the measurements reported in the following. In order not to damage the medium, the pulse energy was limited to about 1 μJ, and the generation of a white-light continuum was normally observed to proceed through the creation of a single light filament. Higher energies involved the breakup of the pulse into multiple filaments, giving rise to highly structured patterns as a result of the spatial interference among the different, and mutually coherent, white-light sources thus created. Although visually interesting, we tried to avoid this effect and all of the measurements described below were performed in the single-filament regime.

3 Results and discussion

In the ideal case, in which the two time-delayed white-light pulses with electric fields $E_1(t)$ and $E_2(t+\tau)$ are perfectly phase-locked, their combined spectrum is easily found as

$$I_t(\omega,\tau) = I_1(\omega) + I_2(\omega) + 2\sqrt{I_1(\omega) I_2(\omega)}\cos(\omega\tau)\,, \qquad (1)$$

which reduces to

$$I_t(\omega,\tau) = 2I(\omega)(1+\cos(\omega\tau)) \qquad (2)$$

in the case of equal supercontinuum intensities $I(\omega) = I_1(\omega) = I_2(\omega)$.

Our objective was then to verify that, when the two white-light pulses are generated by two time-delayed phase-locked laser pulses with reasonably equal efficiencies, a modulated spectrum of the kind expressed in (2) appears and extends all over the visible and near-IR regions.

To this end, we measured the spectra of the pulses after interaction with a 1-mm-thick medium for increasing values of the pump pulse energy. In order not to saturate the detector with the intense fundamental light around 800 nm, colored high- or low-pass filters were used to observe either the visible or the near-IR parts of the spectrum, respectively.

Figure 3 shows the appearance of the white light continuum on the short-wavelength side of the spectrum: at low power, only the red part of the laser spectrum leaking from the filter was visible, with clear fringes due to interference between the two time-delayed pulses. When the pulse energy was increased, visible components appeared at shorter wavelengths, but no interference was observable yet. Only when the pulse energy was increased close to 1 μJ, clear spectral interferences appeared. This is simply explained by the presence of a threshold behavior, combined with a slight imbalance in the peak intensities of the two pump pulses. While at low power neither pulse suffered sufficient self-phase-modulation to significantly broaden its spectrum, at intermediate values one of the two reached the critical power for self-trapping and the intensity in the so-formed filament gave rise to new spectral components in the visible region. Only when the second pulse reached the same threshold was a comparable amount of white-light suddenly coherently generated, allowing the corresponding spectral components of the two pulses to interfere. With a pulse duration of about 30 fs, one can estimate the critical power P_{cr} for self-trapping to be on the order of 20–30 MW, in good agreement with the predictions based on the expression [26]

$$P_{\mathrm{cr}} = \frac{\pi}{8 n_0 n_2} (1.22\lambda)^2 \,, \tag{3}$$

with a nonlinear refraction index n_2 on the order of $10^{-16}\ \mathrm{cm}^2/\mathrm{W}$.

As long as the pulse energy was not increased too much above this single-filament threshold, to a level such that both pulses were able to independently generate stable white-light, the resulting spectrum was always characterized by very stable interference fringes of high contrast extending through all of the visible and down to about 1.1 μm. The pulse peak intensity in the focus under these conditions was estimated to range between 1 and 5 times $10^{12}\ \mathrm{W/cm}^2$ depending on the medium thickness, with a waist radius of about 15 μm. Using thicker transparent materials reduced the pulse energy needed to produce supercontinuum, and the transition to extreme spectral broadening was somewhat smoother. This is probably connected with the fact that, while for thin plates one is forced to reach the threshold for self-focusing and trapping in order to efficiently generate continuum, thicker plates can accumulate larger nonlinear phase modulations upon propagation and can generate white light at lower intensities without the need to undergo drastic self-focusing effects.

By changing the delay τ between the pump pulses it is possible to vary the period of the spectral fringes; in the wavelength domain, the period of the modulation is simply found from (2) to be

$$\delta\lambda = \frac{\lambda^2}{c\tau}. \tag{4}$$

When the pulse delay τ was smaller than the coherence time of the laser pulses (approximately 25 fs), their interference caused strong oscillations in the total yield of supercontinuum, alternating dark regions to situations characterized by structured patterns where multiple filaments are generated. However, for longer delays, the pump pulses no longer temporally overlapped in the medium and both contributed to the generation of phase-locked, white-light pulses. Two examples of the spectra, in the visible and in the near-IR region, corresponding to different time delays between the pulses, are reported in Figs. 4 and 5.

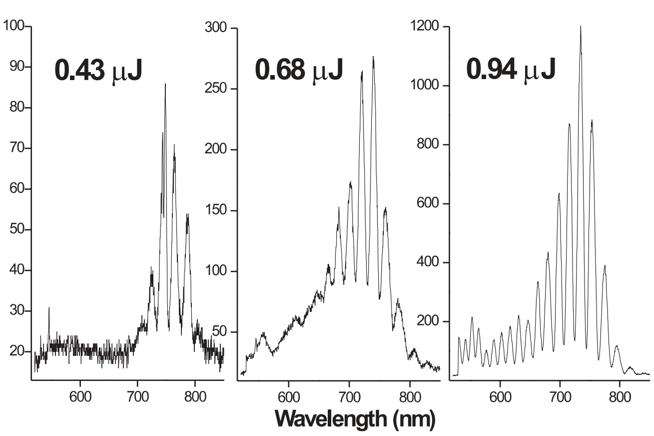

FIGURE 3 Appearance of white-light spectral interference fringes on the visible side of the spectrum in a 1-mm-thick glass plate. In the sequence of spectra measured at different single-pulse energies, one can observe the birth of a single supercontinuum pulse and the appearance of the second phase-locked pulse, which produces a clear sinusoidal modulation in the spectrum

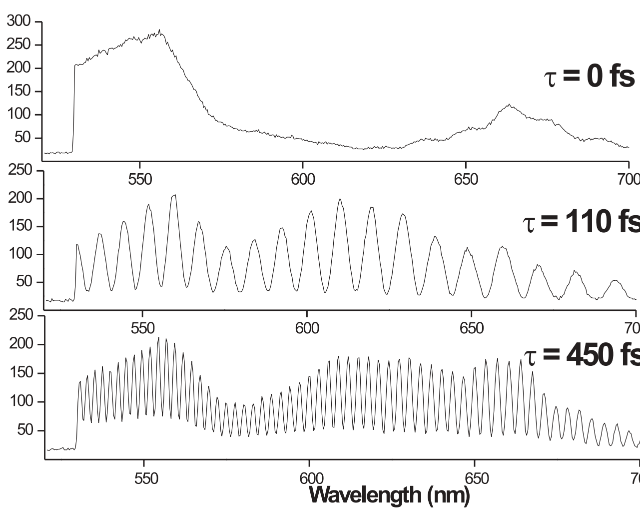

FIGURE 4 Visible portion of the two-pulse supercontinuum spectrum displaying sinusoidal modulations of different periods as a function of the time delay between the pump pulses

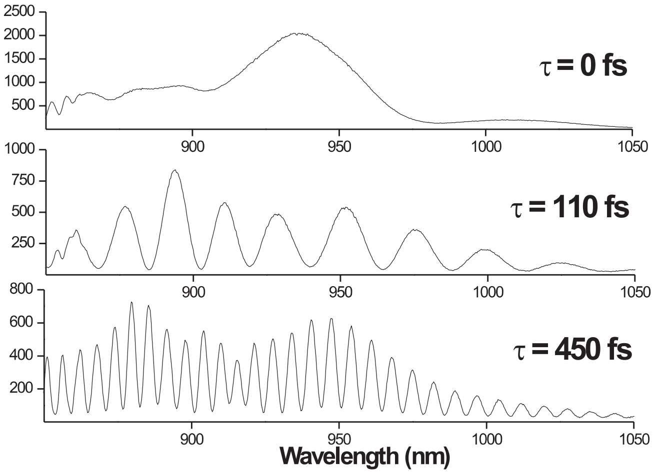

FIGURE 5 Same as Fig. 4 but for the near-IR part of the supercontinuum spectrum

The fringe period measured in the region around 900 nm is also reported in Fig. 6 as a function of the delay. The expected behavior with the inverse of the time separation between the pump pulses is well reproduced by the experimental data.

Increasing the time delay also reduced the visibility V of the observed fringes, defined as

$$V = \frac{I_{max} - I_{min}}{I_{max} + I_{min}} . \tag{5}$$

One immediate explanation for this trend is obviously connected with the finite resolution of the spectrometer: when the period of the spectral modulations $\delta\lambda$ approaches the resolution of the instrument $\Delta\lambda$, it starts to smooth the fringe pattern, finally washing out their visibility. The expression (2) then has to be modified to include the transmission function $F(\omega)$ of the spectral filter and becomes

$$I_t(\omega, \tau) = 2I(\omega)(1 + \tilde{F}(\tau)\cos(\omega\tau)) , \tag{6}$$

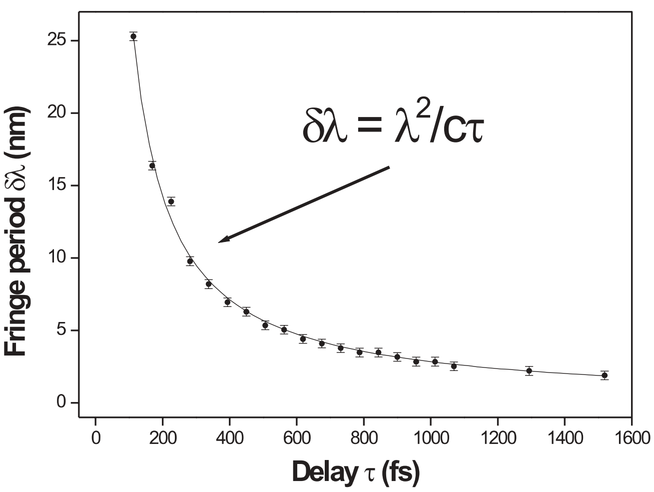

FIGURE 6 Fringe period as a function of the time delay τ between the two laser pulses. *Filled circles* represent the data points and the *solid curve* is a $1/\tau$ fit

where it is assumed that $F(\omega)$ is symmetric and much narrower than the single-pulse spectrum, $\tilde{F}(\tau)$ being its Fourier transform normalized to have $\tilde{F}(0) = 1$. It is evident that $\tilde{F}(\tau)$ corresponds to the fringe contrast defined as in (5).

However, other possible sources of fringe contrast degradation can be identified and essentially derive from a difference in the conversion efficiency of the two laser pulses or by the loss of mutual coherence between the two supercontinua. If the first pulse somehow degrades the medium after its passage, then the second pulse generates much less intense white light and the fringe contrast $V(r)$ decreases as

$$V(r) = \frac{2\sqrt{r}}{1+r} , \tag{7}$$

where $r = I_1(\omega)/I_2(\omega)$ is the ratio of the two supercontinuum intensities. Even without an intensity imbalance between the two white-light pulses, the passage of the first pulse might perturb the phase evolution of the second, contributing to a decrease of the fringe contrast anyway.

In order to discriminate between these sources of contrast decay, we studied the behavior of the fringe visibility as a function of the pulse relative delay for different spectral components. In particular, we determined the decay curves for two regions of the white-light spectrum above and below the laser wavelength, and compared them with the visibility curve for the modulated laser spectrum obtained without interaction with the medium. If the only source of contrast degradation were connected to the limited spectral resolution of the detector, the three curves would be expected to show the same functional behavior (a Gaussian decay curve, if one assumes that the spectrometer has a Gaussian instrumental linewidth) with a decay time scaling quadratically with the wavelength. This is easily explained by the fact that, according to (4), shorter wavelengths display denser fringes than longer wavelengths for the same time delay; if the instrumental spectral resolution $\Delta\lambda$ is constant throughout the spectrum, then the short-wavelength fringes should disappear before the long-wavelength ones. If other sources of contrast decay are present due to the generation process and play a role in the degradation of the visibility, then the functional form of the decay might change and, more importantly, the decay time might become significantly shorter.

The normalized visibility curves are shown in Fig. 7 for the non-interacting laser pulses around 780 nm and for the white-light continuum around 620 and 900 nm. The three curves are well fitted by Gaussian profiles with $1/e$ decay times of 890, 650, and 1300 fs, respectively, in good agreement with the expected quadratic dependence on the wavelength.

The fact that the fringe contrast nicely follows the decay curve expected for the case of purely resolution-limited visibility is further proof that supercontinuum generation does not spoil the phase coherence of the laser pulses, and that this is valid in a wide interval of wavelengths and time delays.

Finally, we tested the effects of pulse energy imbalances on the generation and phase coherence of the white light. We slightly modified the experimental setup by including a quarter-wave plate in one of the arms of the interferometer

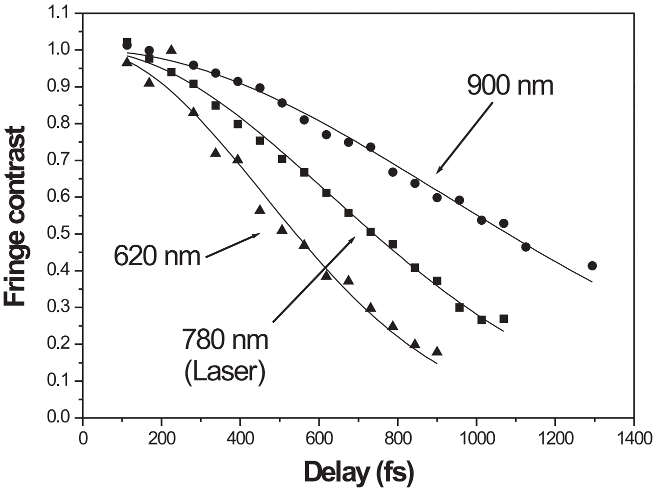

FIGURE 7 Plots of the normalized decay of spectral fringe contrast as a function of the delay between the pulses for the laser (*squares*) and for two regions of the supercontinuum spectrum (*triangles* for 620 nm and *filled circles* for 900 nm). The *solid lines* are Gaussian fits to the experimental data with the decay constant as the only free parameter

and by passing both the output beams through a cube polarizer. The waveplate was mounted on a computer-controlled precision rotator to allow for smooth and repeatable movements. A double passage of the pulse through the $\lambda/4$ plate has the net effect of a single passage through a $\lambda/2$ plate that can thus rotate the linear polarization of the laser. With such a setup it was possible to continuously vary the intensity of one of the two pump pulses and verify the effects on the spectral fringe patterns.

We found that changing the energy of one of the two pulses over a relatively wide interval from 0.2 μJ, the threshold for continuum generation in a 5-mm-thick plate, to about 0.4 μJ, corresponding to the energy of the other pulse, the fringe contrast varied in a smooth way from 0 to 1 as illustrated in Fig. 8 for two different wavelength regions. It is also evident from the figure that the two spectral components present a different behavior, with the zone close to 650 nm surviving the higher intensity imbalance. It is however interesting to note that, close to the balanced condition, both spectral components could stand an intensity variation in the generating pulse as high as 30% without substantially degrading the fringe contrast.

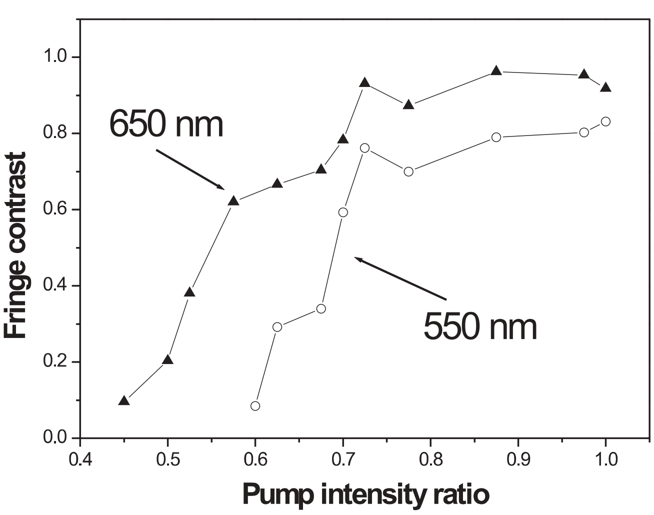

FIGURE 8 Fringe contrast as a function of the pump pulse energy imbalance for two spectral regions of the supercontinuum: *triangles* correspond to 650 nm while *circles* to 550 nm. *Solid lines* connecting the experimental points are just guides for the eye

Changing the intensity of one beam also produced noticeable effects in the positions of the fringes. For an energy variation of 0.5 μJ, roughly corresponding to a variation ΔI of $3 \times 10^{12}\ \mathrm{W/cm^2}$ for the pulse peak intensity in the focus, we observed a shift of about 6 fringes in the spectral interference pattern around 600 nm. The fringe shift introduced while increasing the pulse intensity in one arm could be compensated for by increasing the path length in the other interferometer arm. This is consistent with the fact that an increase of pulse intensity corresponds to an increase of the effective optical path in the case of a positive nonlinear refraction index n_2.

In the above case, one easily verifies that this fringe shift is compatible with that calculated from

$$\Delta\varphi = \frac{2\pi}{\lambda} n_2 \Delta I L \tag{8}$$

for propagation in a medium of thickness $L \approx 5$ mm and nonlinear index $n_2 \approx 10^{-16}\ \mathrm{cm^2/W}$ in the case of a similar intensity variation. From the above considerations one can conclude that moderate intensity fluctuations do not seem to affect the position and visibility of supercontinuum fringes too much, leading to relatively stable modulation patterns even in the presence of energy fluctuations of several percent.

4 Conclusions

We have verified the preservation of mutual phase coherence between supercontinuum pulses generated in bulk media by two phase-locked laser pulses in a collinear configuration. Although this is the fundamental process that is the basis of the recent technique of frequency comb generation in photonic fibers with mode-locked lasers, the equivalent mechanism in bulk media takes place at much higher intensities and the nonlinear processes involved are far less smooth and controllable. By observing clear and stable spectral interference fringes throughout the spectrum ranging from 500 to about 1100 nm, we have demonstrated the robustness of the white-light phase characteristics, which are preserved even in the presence of the strong nonlinearities involved in the generation process. The mutual coherence already observed in previous work [13, 21] between spatially separated sources of supercontinuum does not seem to be spoiled in the collinear configuration, which relies on generation from the same region of the medium and suffers from possible deleterious effects due to interference between the pump pulses. We have also studied the effects of pump intensity fluctuations and have found that, over a relatively large range, they do not spoil the interference patterns, thus indicating a substantial tolerance of the mutual phase coherence to moderate instabilities.

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