Phase-locked white-light continuum pulses: toward a universal optical frequency-comb synthesizer

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We demonstrate that two white-light continuum pulses that are independently generated by phase-locked ultrashort laser pulses are locked in phase and show surprisingly clear and stable Young interference fringes. The experiment shows that the two generated continua emit essentially in phase and that random phase jitter can remain negligible. This result is not only of interest for studies of nonlinear field–matter interactions but also suggests that such white-light continuum pulses can be used to realize a broad frequency comb for absolute frequency measurements from the IR to the UV.

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Recent experiments[1–4] have shown that trains of ultrashort laser pulses are a powerful tool for measuring optical frequencies with very high precision. The comb of frequencies (spaced by a known pulse-repetition rate) that constitutes the spectrum of a mode-locked laser can be used as a ruler to measure frequency differences up to tens of terahertz with an accuracy of a few parts in 10^{17}. The largest frequency gap that can be bridged in such a way is determined by the bandwidth of the laser radiation and, for transform-limited pulses, the shorter the pulse, the larger the frequency interval available.

A commercial Ti:sapphire-based mode-locked laser with a pulse duration of ~75 fs was used in the research reported in Ref. 2 to measure a frequency gap of ~18 THz and determine the absolute optical frequency of the cesium D_1 line at 895 nm to parts in 10^{10}. A comb that was broadened to more than 50 THz by self-phase modulation in a nonlinear optical fiber was used in a recent absolute frequency measurement of the hydrogen $1S-2S$ interval.[3] A similar technique[4] permitted measurement of the 104-THz frequency gap between two cw lasers at 1064 and 778 nm with a relative uncertainty of $\sim 10^{-11}$.

To bridge even larger frequency gaps and eventually build a universal optical frequency meter across the whole visible spectrum and extending into the near-UV and the IR parts of the spectrum will require even broader combs.

Once a frequency comb spanning more than an octave becomes available, the absolute optical frequencies of the comb will be attainable directly from measurements of the pulse-repetition rate in the rf domain in a single step. One can either bridge the gap between a laser frequency and its second harmonic[3] or observe the beat note between the blue end and the second harmonic of the red end of the same frequency comb.

One possible way to achieve such extreme spectral broadening relatively simply is through white-light continuum generation.[5,6] Provided that the laser pulses are intense enough, focusing them into a suitable transparent material results in the generation of a white-light continuum that contains wavelengths ranging from the IR to the near UV. Although continuum generation is a complex issue involving changes in the temporal and spatial beam characteristics, the dominant process and the starting mechanism leading to spectral superbroadening is the self-phase modulation of the pulse, which is due to the intensity-dependent refractive index of the medium. In any material with a third-order nonlinear susceptibility $\chi^{(3)}$ and an instantaneous response, the refractive index depends on the intensity $I(t)$ of the propagating field:

$$n(t) = n_0 + n_2 I(t)\,, \tag{1}$$

so, after propagation through a short length L of such a medium, the field at a carrier frequency ω_0 experiences a time-dependent shift in the instantaneous frequency that is equal to

$$\Delta\omega(t) = -\omega_0 n_2 L/c\,\frac{\mathrm{d}I(t)}{\mathrm{d}t}\,, \tag{2}$$

resulting in red detuning at the leading edge of the pulse and blue detuning at the trailing edge. A 100-fs pulse focused to a peak intensity of 10^{14} W/cm^2 in a 1-mm-thick medium with a typical nonlinear index coefficient of 10^{-16} cm^2/W will give rise to a continuum pulse with a frequency excursion that is comparable to the frequency of the carrier itself.

The process of self-phase modulation alone, however, is not sufficient to characterize fully the phenomenon of white-light generation, and a number of other linear and nonlinear effects play a role as well. The self-focusing effect alters the transverse spatial distribution and modifies the intensity of the beam along its path; parametric four-photon mixing, stimulated Raman and Brillouin scattering, and shock-wave formation may contribute to the distortion of the pulse shape and to the broadening of the spectrum; and, finally, all these nonlinear interactions must be considered in combination with group-velocity dispersion. The generation of the continuum is, then, the result of very complex interplay among competing processes, and the exact characteristics of the output beam appear to be strongly dependent on the exact initial conditions of the interaction and are hardly predictable. In particular, one is led to expect that the white-light pulses produced by phase-locked pump pulses have lost any precise phase relationship in the generation process. Considering that phase coherence among successive pulses in the train is an essential ingredient for the generation of a broadband frequency comb, such white-light pulses may, at first glance, appear inadequate for this purpose.

We have decided to test the phase coherence of the generated continuum pulses by performing an experiment related to Young's double-slit experience: Here two white-light pulses are generated independently at different positions in a transparent medium by two phase-locked laser pulses (see the inset of Fig. 1). Our results are rather intriguing: When the two pump pulses are adjusted for zero relative delay, the two white-light continua that they generate independently show clear and stable interference fringes, indicating that we are dealing with highly phase-correlated secondary sources.

We emphasize that there is a substantial difference between this approach and a simple Young- or Michelson-type experiment: In such cases two spatial portions of the same beam or two time-delayed replicas of the same pulse are recombined to give interference. In our experiment, however, the interference fringes appear because of the spatiotemporal superposition of two white-light pulses that are independently generated in two separate positions of the medium. For the reasons discussed above, such pulses might be expected to be highly uncorrelated.

The experimental setup is particularly simple and is shown schematically in Fig. 1. It is similar to one that was previously used to measure the temporal coherence of ultrashort extreme-UV pulses produced in the process of high-order harmonic generation.[7] The output from an amplified mode-locked Ti:sapphire laser delivering 0.5-mJ, 100-fs pulses at a repetition rate of 1 kHz is fed, after proper attenuation, to a Michelson interferometer, which is slightly misaligned so that the outgoing beams are not perfectly parallel to each other and are focused in two separate regions by a 150-mm focal-length lens. For most of our experiments a 2-mm-thick CaF_2 plate has been placed in the focal plane. Both beams independently produce a white-light pulse that emerges from the interaction zone, and, after the two diverging continua have propagated in air for some distance, they finally overlap on a screen, where interference fringes, if any, are detected.

When the pulse energy is not too far above the threshold for self-focusing and the time delay between the pump pulses is properly adjusted, clear and stable white interference fringes appear on the screen. A snapshot of the observed interference pattern is shown in Fig. 2. The only observed instabilities do not seem to depend on the process of generation but are due to residual mechanical vibrations in the interferometer and, in an exposure time of ~2 s, do not degrade the fringe contrast significantly. By changing the tilt angle of one mirror in the interferometer, we could vary the separation of the two focal spots in the medium (from approximately 50 to 200 μm) and consequently vary the fringe spacing. Note that in any case the two sources of white light are always well separated, compared with a measured waist radius of less than 15 μm in the focal plane.

Similar fringe patterns have been observed by use of different materials as the nonlinear medium: Quartz plates and plain glass microscope slides have proved effective for producing stable interference, as have

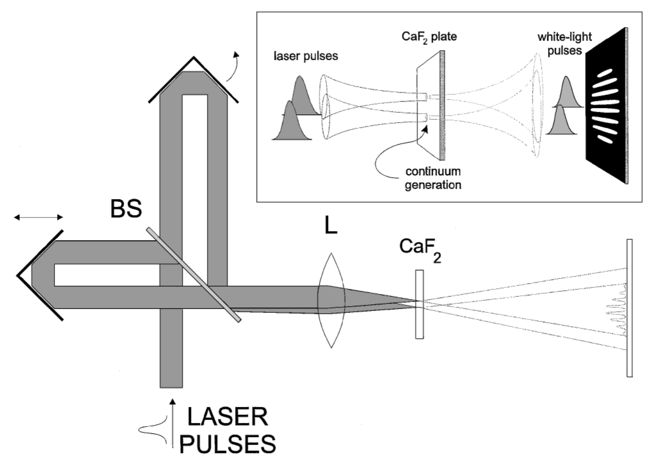

Fig. 1. Experimental setup for testing the phase lock between two white-light continuum pulses. The IR pulses from the laser are split by a 50% beam splitter (BS) and focused with a variable relative delay by lens L onto a thin CaF_2 plate. Interference fringes between the two emerging continua are detected on a screen in the far field.

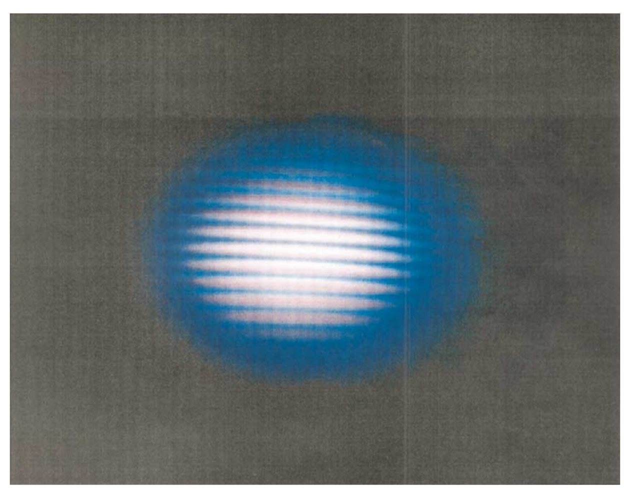

Fig. 2. White-light fringes resulting from the interference of the two continua generated by the two phase-locked IR laser pulses when the relative delay is properly adjusted to zero.

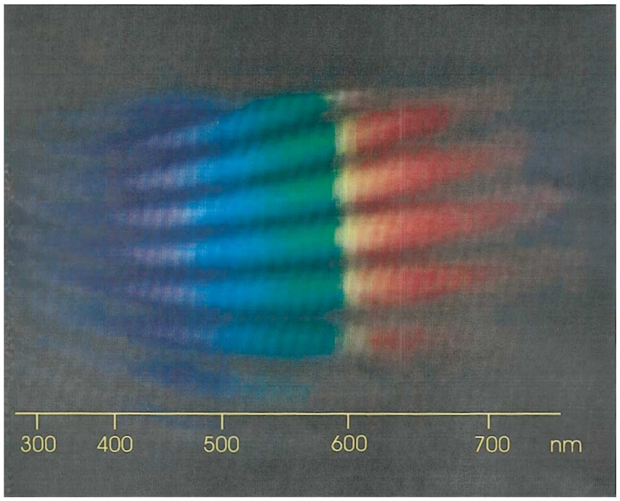

Fig. 3. Spectrally dispersed white-light fringes. Clear and well-defined fringes indicate that a stable phase relationship is conserved across all the generated visible spectrum.

a water cell and a 5-mm-thick Plexiglas plate. In general, any transparent material has produced white-light continua with various degrees of efficiency, and interference fringes have always clearly appeared, showing that the phenomenon is not dependent on the material.

So as not to damage the crystals by destructive optical breakdown, and to avoid multiple filamentation in the medium, we limit the energy of each pulse coming from the interferometer to less than 3 μJ. Under such conditions, a single filament is generally created by each focused beam in the transparent material, with an estimated peak intensity of the order of 10^{13} W/cm^2. Nonetheless, we have also observed that, even in the case of multiple filaments, and depending on their number, more- or less-complicated interference structures appear, indicating that the mutual coherence among the resulting continua is conserved.

To investigate whether the two white-light continuum pulses are mutually phase coherent over the entire visible spectrum, we have spectrally dispersed the interference pattern with a planar diffraction grating and a cylindrical lens: When the two focal spots are aligned in a direction parallel to the grating grooves, the interference fringes are parallel to the direction of the spectral dispersion and can be followed as they change color. Also in this case it is possible to produce stable fringe patterns of high contrast over the entire spectrum, as shown in Fig. 3.

We have also qualitatively investigated the effect of changing the relative intensities of the two primary pulses. By partly blocking one of the two beams until the power is reduced by approximately half, we measure an intensity-dependent pulse advance of the order of 25 fs, or a phase shift of ~100 rad at 500 nm, in a 2-mm-thick CaF_2 plate. This result implies that relative phase changes can remain less than 1 rad as long as the relative pulse intensities are balanced within 0.5%.

Concerning the possible applications of these findings to the realization of what we have called a universal optical frequency-comb synthesizer for frequency measurements across the whole visible range, we must note that most existing sources of white-light continuum pulses rely on amplified lasers to reach the intensities required for spectral broadening. Such sources necessarily have a limited repetition rate (typically 1 to 5 kHz, with some systems reaching hundreds of kilohertz), and their spectrum consists of very narrowly spaced modes. To make a frequency-comb measurement one has to know exactly the integer number of modes between the frequency to be measured and the reference frequency, and it is then necessary to have a good preliminary knowledge of the unknown frequency gap, with an uncertainty less than the separation between two successive modes. Satisfying this condition is conceivable only with sufficiently high repetition rates of at least some tens of megahertz. Ideally the mode separation should be of the order of hundreds of megahertz, so that a simple wave meter is sufficient for accurately determining the frequency gap and unambiguously counting the number of spanned modes. Although it was not possible to generate a white-light continuum with the low peak intensities that are available with the mode-locked systems developed so far, recent reports[8] demonstrated that photonic crystal fibers allow the generation of a supercontinuum directly from the output of a mode-locked femtosecond laser with pulse energies of a few nanojoules or less. The experiments reported here support recent observations[3] that supercontinuum generation in such fibers can be the key to the realization of a universal frequency synthesizer.

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