Laser and Particle Beams (2001), *19*, 157–162. Printed in the USA.

Generation and applications of phase-locked white-light continuum pulses

MARCO BELLINI[1,2,3] AND THEODOR W. HÄNSCH[2,3,4,5]
[1]Istituto Nazionale di Ottica Applicata (INOA), L.go E. Fermi, 6, 50125, Florence, Italy
[2]European Laboratory for Non Linear Spectroscopy (LENS), L.go E. Fermi, 2, 50125, Florence, Italy
[3]Istituto Nazionale di Fisica della Materia (INFM), L.go E. Fermi, 2, 50125, Florence, Italy
[4]Department of Physics, University of Florence, L.go E. Fermi, 2, 50125, Florence, Italy
[5]Max-Planck-Institut für Quantenoptik, P.O. Box 1513, D-85740 Garching, Germany

(RECEIVED 9 November 2000; ACCEPTED 5 February 2001)

Abstract

We report on an experiment demonstrating the high degree of mutual phase coherence between two white-light continuum pulses independently generated by phase-locked ultrashort laser pulses. We show that these secondary pulses are locked in phase and exhibit surprisingly clear and stable Young's interference fringes when overlapped on a screen in the far field. We discuss the use of such phase-locked continuum pulses in the realization of a new kind of ultrabroad frequency comb for absolute frequency measurements throughout the visible spectrum.

1. INTRODUCTION

The rapid progress in femtosecond pulse generation has made it possible to generate light pulses with durations down to a few optical cycles and with intensities able to drive highly nonlinear phenomena (see Brabec & Krausz, 2000, and references therein). The extreme temporal resolution of such short pulses has been mainly exploited to investigate ultrafast dynamics on the femtosecond time scale (Cerullo *et al.*, 1999) or, on the other hand, their high peak intensity has allowed the generation of coherent radiation in new, and often hard to reach, spectral ranges. The generation of high-order harmonics in the deep ultraviolet and in the soft X-ray regions is just one of the most impressive applications of this new kind of laser sources (for a recent review see Salières *et al.*, 1999).

Recent experiments (Udem *et al.*, 1999; Diddams *et al.*, 2000*a*; Reichert *et al.*, 2000) have shown, however, that trains of ultrashort laser pulses can also be a powerful tool to measure optical frequencies with very high precision.

At first glance it does not seem to make sense to perform high-precision spectral measurements with ultrashort laser pulses, due to their intrinsic broad bandwidth (see Fig. 1a). If a narrow spectral feature (such as the absorption line corresponding to some long-lived atomic transition) is to be analyzed, the resulting spectrum is always the convolution between such a narrow line and the much broader instrumental profile. This unavoidably washes away any fine structure and gives an uncertainty of the order of the laser bandwidth on any possible spectral measurement.

However, when two or more time-delayed and phase-locked ultrashort laser pulses are used, the situation changes and higher spectral resolutions can be achieved.

In the simple case of a pair of phase-locked pulses, the broad (of the order of the inverse of the pulse duration), bell-shaped spectrum corresponding to the single laser pulse acquires a sinusoidal modulation with a period given by the inverse of the time delay between the pulses (see Fig. 1b). The new spectral resolution of the system is set by this period of modulation and, in principle, it can be improved just by imposing longer and longer delays between the pulse pair while maintaining their phase lock. While the wide spectral bandwidth of the pulses still makes it possible to record survey-type optical spectra, the peak intensity of the spectrally filtered light is independent of the time delay and is still capable of exciting multiphoton transitions or of producing high harmonics when focused in a gas jet.

This kind of Fourier spectroscopy with ultrashort laser pulses has been recently demonstrated for the study of two-photon atomic transitions (Bellini *et al.*, 1997) and we are currently working to extend this technique to perform high-resolution spectroscopy in the XUV, with phase-locked high-order harmonic pulses.

Address correspondence and reprint requests to: Marco Bellini, Istituto Nazionale di Ottica Applicata, L.go E. Fermi, 6, I-50125 Florence, Italy. E-mail: bellini@ino.it

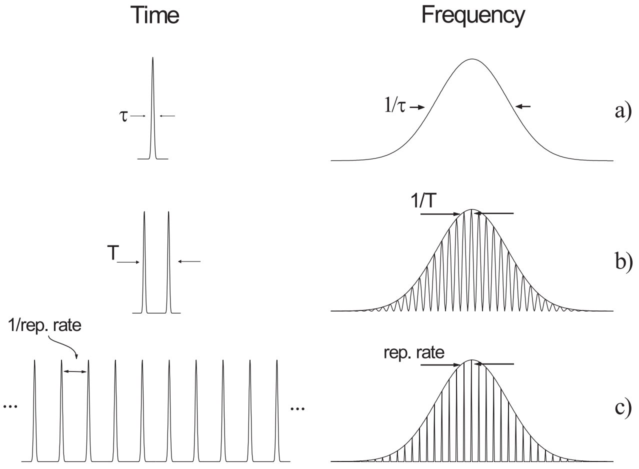

Fig. 1. Schematic comparison among the spectra corresponding to different temporal pulse sequences: (a) Single pulse; (b) Pair of time-delayed and phase-locked pulses; (c) Train of phase-locked pulses with a constant delay. The last configuration corresponds to the time evolution and spectrum of a mode-locked laser.

Increasing the number of phase-locked and constantly time-delayed pulses in the sequence generates sharper and sharper peaks in the corresponding spectrum, until a precise set of equally spaced modes resembling the teeth of a comb is produced. The spacing between the modes (the teeth of the comb) is just the inverse of the time delay between the pulses.

A mode-locked laser is a particularly simple generator of an ideally infinite sequence of time-delayed pulses with a well-defined phase relationship. Not surprisingly, its spectrum (given by the set of equally spaced longitudinal modes of the cavity) is a broad comb of frequencies with a total width given by the inverse of the pulse duration and a mode separation equal to the measurable and controllable pulse repetition rate (see Fig. 1c).

The spectral comb generated by a mode-locked laser can thus be used as a precise ruler to measure large frequency differences with high accuracy. If a known optical reference frequency ν_{ref} is available and one mode of the comb is locked to it, the frequency of all other modes is unambiguously determined by $\nu_x = \nu_{ref} + N\nu_{rep}$, where N is an integer and ν_{rep} is the known repetition rate. The frequency of another laser falling within the bandwidth of the comb can then be obtained by comparing it to the nearest mode of the comb itself.

To eliminate ambiguities in the determination of the integer number of spanned modes N, it is necessary to possess a good preliminary knowledge of the unknown frequency gap with an uncertainty less than the separation between two successive modes. This is only conceivable 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 wavemeter is sufficient to accurately determine the frequency gap and unambiguously count the number of spanned modes.

The largest frequency gap that can be bridged with this comb technique is determined by the bandwidth of the mode-locked laser producing the comb and, for transform-limited pulses, the shorter the pulse, the larger is the frequency interval available.

A commercial, Ti:sapphire-based, mode-locked laser with a pulse duration of about 75 fs was used by Udem *et al.* (1999) to measure a frequency gap of about 18 THz and determine the absolute optical frequency of the cesium D_1 line at 895 nm to parts in 10^{10}. A comb broadened to more than 50 THz by self-phase-modulation in a nonlinear optical fiber has been used in a recent absolute frequency measurement of the hydrogen 1*S*-2*S* interval (Reichert *et al.*, 2000). A similar technique (Diddams *et al.*, 2000*a*) allowed the measurement of the 104 THz frequency gap between two cw lasers at 1064 and 778 nm with a relative uncertainty of about 10^{-11}.

To bridge even larger frequency gaps and eventually build a universal optical frequency-meter across the whole visible spectrum and extending in the near UV and infrared parts of the spectrum, even broader combs are needed. Once a frequency comb spanning more than one optical octave is available, absolute optical frequencies can be obtained directly by measuring the frequency gap between a laser frequency and its second harmonic, if they both fall under the broad comb span. A measurement of the pulse repetition rate in the radio-frequency domain then allows one to bridge the gap between the cesium frequency standard and the optical domain in a single step (Diddams *et al.*, 2000*b*).

The process of white-light continuum generation (Fork *et al.*, 1983; Corkum *et al.*, 1986) may provide an efficient way to achieve such an extreme spectral broadening in a relatively simple way. Provided that the laser pulses are intense enough, focusing them into a suitable transparent material results in the generation of a white-light continuum, containing wavelengths ranging from the IR to the near UV. A white-light comb can be generated if a train of phase-locked infrared pulses from a mode-locked laser is used to generate a train of phase-locked, white-light pulses after the interaction with the medium.

There are two main obstacles to the realization of such a wide comb: The first is that there is no guarantee about the preservation of the phase coherence between successive pulses in the output train, and the second depends on the limited pulse energy of current mode-locked systems that prevents the generation of a supercontinuum at high repetition rates.

Self-phase-modulation of the pulse due to an intensity-dependent refractive index of the medium is the dominant process and the starting mechanism leading to spectral superbroadening. After propagation through a short length L of a medium with a nonlinear refractive index n_2, the field at a carrier frequency ω_0 experiences a time-dependent shift in the instantaneous frequency equal to

$$\Delta\omega(t) = -\omega_0 n_2 L/c \, \frac{dI(t)}{dt}, \tag{1}$$

which results in a red detuning at the leading edge of the pulse and a blue one at the trailing edge.

Self-phase-modulation alone, however, is not sufficient to fully characterize the phenomenon of white-light generation and a number of other linear and nonlinear effects play a role as well. Self-focusing 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 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 a very complex interplay between competing processes and the exact characteristics of the output beam appear strongly dependent on the exact initial conditions of the interaction and 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 the 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 a first glance, appear inadequate for this purpose.

The main result of this paper is the test of the mutual phase coherence of pairs of continuum pulses with 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 inset of Fig. 2).

Our results are rather intriguing: When the two pump pulses are adjusted for equal intensity and zero relative delay, the two white-light continua that they generate independently show surprisingly clear and stable interference fringes, indicating that we are dealing with highly phase-correlated secondary sources.

We want to emphasize that there is a substantial difference between this and a simple Young's or Michelson's 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, on the contrary, the interference fringes appear because of the spatio-temporal superposition of two white-light pulses independently generated in two separate positions of the medium. For the reasons discussed above, such pulses might be expected to be highly uncorrelated.

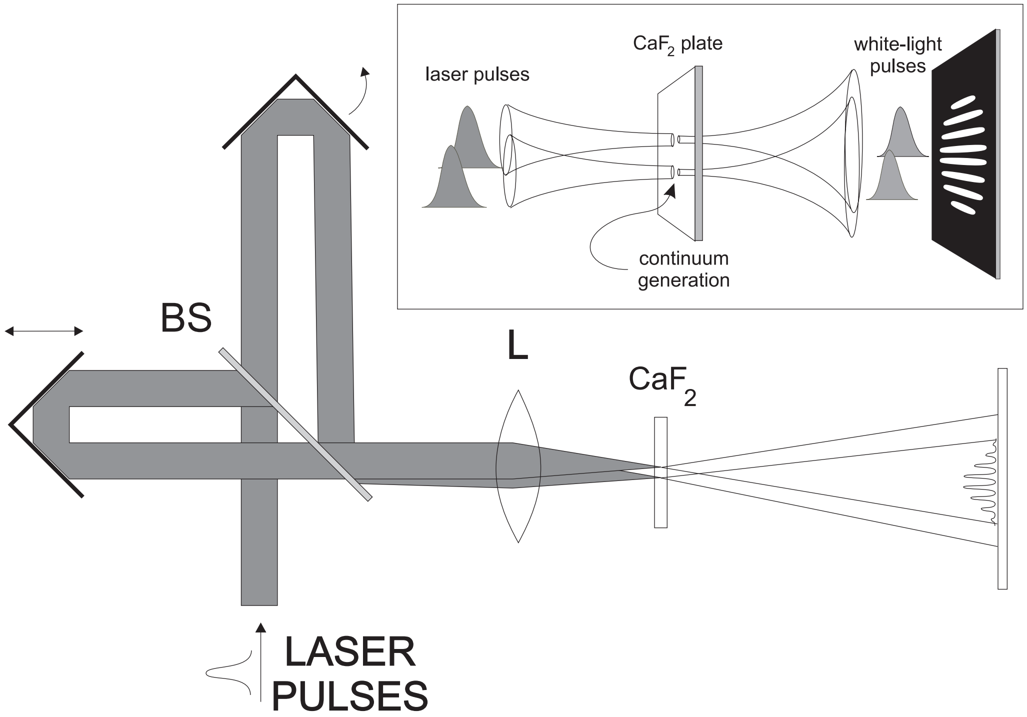

Fig. 2. Experimental setup for the test of the phase lock between two white-light continuum pulses. A 50% beamsplitter (BS) splits the IR pulses from the laser and the lens L focuses them in a thin CaF$_2$ plate with a variable relative delay. Interference fringes between the two emerging continua are detected on a screen in the far field.

Note that, while the light from any source, even very incoherent ones (like a plasma or a bulb), produces interference if sent through a conventional Michelson interferometer which compares two time-delayed portions of the same pulse (of course, for delays shorter than the coherence time), two such uncorrelated sources would not show any kind of interference fringe in the experiment that we propose.

2. EXPERIMENT

The experiment setup (see Fig. 2) is similar to the one previously used by our group to measure the temporal coherence of ultrashort XUV pulses produced in the process of high-order harmonic generation (Bellini *et al.*, 1998).

We use an amplified mode-locked Ti:sapphire laser system, delivering 0.5-mJ, 100-fs pulses at a repetition rate of 1 kHz. After proper attenuation by a half-wave plate and a polarizer, the laser pulses are sent through a Michelson interferometer which is slightly misaligned so that the outgoing beams are not perfectly parallel to each other. The pulses are thus focused in two separate regions of the nonlinear material by a 150 mm focal length lens. By changing the tilt angle of one mirror in the interferometer, we can vary the separation of the two focal spots in the medium (from about 50 to 200 μm). It is worth mentioning that in any case the two secondary sources are always well separated, if compared to a measured waist radius of less than 15 μm in the focus.

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 emerging 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.

3. RESULTS AND DISCUSSION

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. Of course, due to the extremely broad spectrum connected to these white-light pulses, their coherence time is very short and the interference fringes are observable only in a limited delay range (of the order of a few femtoseconds) around zero. Also at zero delay, just a few central fringes appear really white and, moving towards the outer regions of the pattern, the different colors (corresponding to different fringe spacings) start to separate until no more white light is observable.

A snapshot of the observed interference pattern is shown in Figure 3. The only instabilities are due to residual mechanical vibrations in the interferometer but, in an exposure time of several seconds, they do not degrade the fringe contrast significantly. This shows that the phase lock is not only preserved on a shot-to-shot basis, but that a constant phase relationship is maintained over at least thousands of shots. Also note that, in order not to damage the material, we constantly move it so that different areas of the CaF_2 plate interact with the laser pulses at each shot. If small inhomogeneities are present on the surface or if there is some intensity fluctuation on the laser pulses, their effect on the mutual coherence of the two white light beams must be completely negligible.

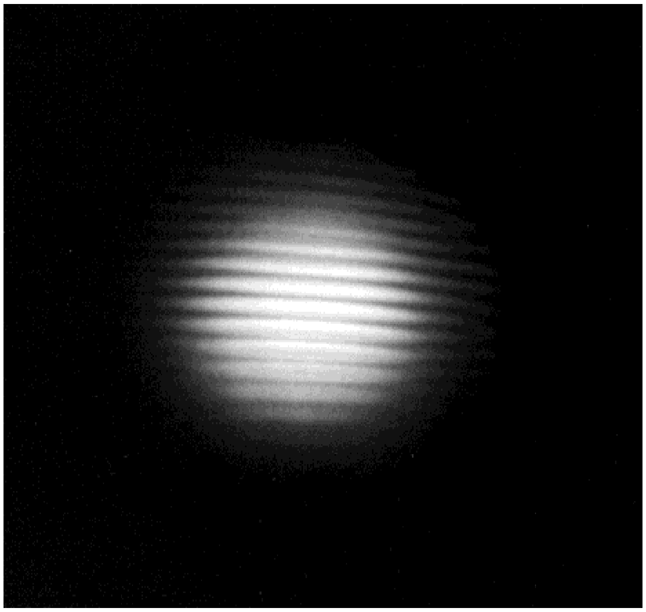

Fig. 3. White-light interference fringes generated when the relative delay between the two phase-locked pump pulses is properly adjusted to zero.

Fringe patterns of similar stability and contrast have been observed using different materials as the nonlinear medium: Quartz plates and plain glass microscope slides have proved effective to produce a stable interference, as well as a water cell and a 5-mm-thick Plexiglas plate. In general, any transparent material present in the laboratory has proved able to produce white-light continua at various degrees of efficiency, and interference fringes have always clearly appeared, showing that the phenomenon is not dependent on the material.

In order not to damage the crystals by destructive optical breakdown and to avoid multiple filamentation in the medium, the energy of each pulse coming from the interferometer is limited 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 between 10^{12} and 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.

The effect of changing the relative intensities of the two primary pulses has also been qualitatively investigated.

Again, considering the complex interplay of linear and nonlinear phenomena involved in the generation of the white light, it might be expected that a slight unbalancing of the pump intensities can lead to a complete loss of the mutual coherence between the two continua.

The unbalance in the intensities has been obtained by partly blocking one of the two beams until its power is approximately cut in half. Of course this also results in a lower efficiency in the generation of the white light, and the different intensities in the two output beams clearly degrade the fringe contrast.

Apart from this, nice and stable fringe patterns have been observed also in this case, with a measured intensity-dependent pulse advance of the order of 25 fs, or a phase shift of about 100 rad at 500 nm, in our 2-mm-thick CaF_2 plate. This also implies that relative phase changes can remain below 1 rad as long as the relative pulse intensities are balanced within 0.5%.

The fact that the pulse is seen to advance in time when the intensity of the pump is diminished can be easily understood recalling that the nonlinear index coefficient n_2 is positive and decreasing the intensity is equivalent to decreasing the effective optical path of the pulse inside the material. The observed temporal shifts are compatible with the estimated value of n_2 for CaF_2.

An even more visual demonstration of the effective phase-lock over the whole visible spectrum has been obtained by recording the spectrally dispersed interference pattern.

A planar diffraction grating and a cylindrical lens have been used, with the two focal spots aligned in a direction parallel to the grating grooves. In such a configuration, 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 has been possible to produce stable fringe patterns of high contrast over the entire visible spectrum, as shown in Figure 4. This is a clear indication that the two continua are mutually phase-coherent over the entire visible spectrum.

The need for a precise determination of the zero delay between the two pump pulses to observe fringes is much less

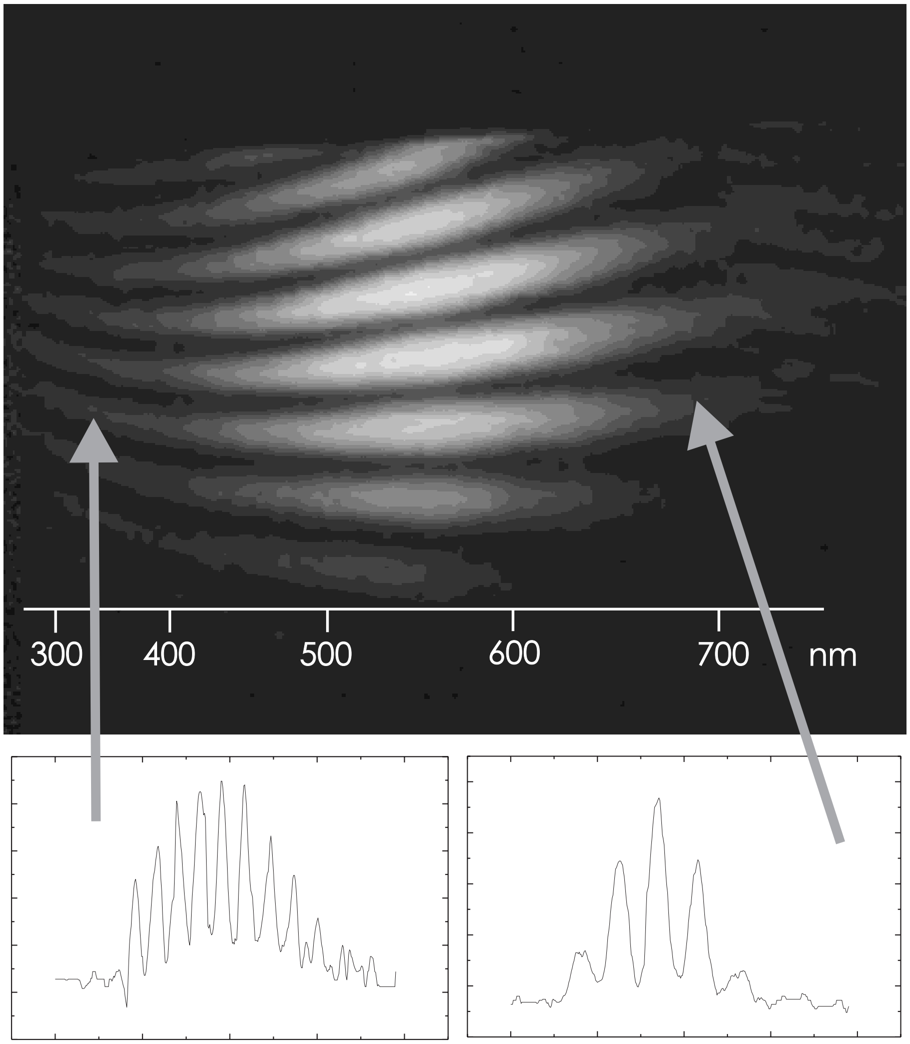

Fig. 4. Spectrally dispersed white-light fringes (note that in the original picture the color of the fringes changes from violet to red when moving from the left to the right side of the screen). Clear and well-defined fringes indicate that a stable phase relationship is conserved across all the generated visible spectrum. Also reported are two vertical lineouts showing that more than one optical octave is spanned by the continuum: The fringes in the right graph have a period which is more than twice as large that of the fringes in the left graph.

stringent in this case. In fact, the spectral dispersion operated by the grating effectively increases the coherence time of each portion of the spectrum and only a global tilt in the fringe pattern is observed when changing the delay up to some tens of femtoseconds.

Also the second obstacle to the realization of an ultrabroad comb for absolute frequency measurements across the whole visible range, that is, the need for a high repetition rate, only achievable with mode-locked systems of low peak intensity, has been recently overcome. It has been demonstrated (Ranka *et al.*, 2000) that new *photonic crystal fibers* allow the generation of a supercontinuum already with pulse energies of a few nanojoules or below.

Thanks to our observations and to the development of these special fibers, a new kind of universal optical frequency synthesizer is now available and, thanks to its compactness and ease of use (compared to the complexity of existing frequency chains), it will rapidly revolutionize the whole field of frequency metrology, with a wide range of applications, from accurate tests of QED to the search of slight asymmetries between matter and antimatter.

ACKNOWLEDGMENTS

This work was supported by the Italian MURST and by the European Community Contract ERB FMGE CT 950017.

REFERENCES

Bellini, M., Bartoli, A. & Hänsch, T.W. (1997). *Opt. Lett.* **22**, 540.

Bellini, M., Lyngå, C., Tozzi, A., Gaarde, M.B., Hänsch, T.W., L'Huillier, A. & Wahlström, C.G. (1998). *Phys. Rev. Lett.* **81**, 297.

Brabec, T. & Krausz, F. (2000). *Rev. Mod. Phys.* **72**, 545.

Cerullo, G., Nisoli, M., Stagira, S., De Silvestri, S., Tempea, G., Krausz, F. & Ferencz, K. (1999). *Opt. Lett.* **24**, 1529.

Corkum, P.B., Rolland C. & Srinivasan-Rao, T. (1986). *Phys. Rev. Lett.* **57**, 2268.

Diddams, S.A., Jones, D.J., Ma, L.-S., Cundiff, S.T. & Hall, J.L. (2000*a*). *Opt. Lett.* **25**, 186.

Diddams, S.A., Jones, D.J., Ye, J., Cundiff, S.T., Hall, J.L., Ranka, J.K., Windeler, R.S., Holzwarth, R., Udem, T. & Hänsch, T.W. (2000*b*). *Phys. Rev. Lett.* **84**, 5102.

Fork, R.L., Shank, C.V., Hirlimann, C. & Yen, R. (1983). *Opt. Lett.* **8**, 1.

Ranka, J.K., Windeler, R.S. & Stentz, A.J. (2000). *Opt. Lett.* **25**, 25.

Reichert, J., Niering, M., Holzwarth, R., Weitz, M., Udem, T. & Hänsch, T.W. (2000). *Phys. Rev. Lett.* **84**, 3232.

Salières, P., L'Huillier, A., Antoine, P. & Lewenstein, M. (1999). *Adv. At. Mol. Opt. Phys.* **41**, 83.

Udem, T., Reichert, J., Holzwarth, R. & Hänsch, T.W. (1999). *Phys. Rev. Lett.* **82**, 3568.