High-order harmonics and white light: looking for fringes and finding much more

Marco Bellini

Istituto Nazionale di Ottica Applicata, LENS and INFM,
Largo E. Fermi 6, I-50125 Firenze, ITALY
bellini@ino.it

1 Introduction

Complex and expensive experimental systems are not always necessary to perform interesting research and to shed light on novel physical phenomena. Very simple experiments often provide great food for thought and can sometimes give new, deep, physical insight when applied in different contexts from those they were initially thought for.

It certainly takes an unusual dose of ingenuity and creativity to think of such simple but decisive experiments but, perhaps more importantly, it takes an exceptionally open mind to look at the results of these experiments from different, and apparently distant, points of view, in order to learn something completely new.

Since 1996, I have been lucky enough to work with Ted Hänsch in the frame of his collaboration with the University of Firenze. During this time, I have had the opportunity to appreciate such rare qualities in him and to be inspired by his enthusiasm and curiosity towards the beauty and the challenges of physics.

In the following I will give a short description of a couple of key experiments we have been performing together at LENS, the European Laboratory for Nonlinear Spectroscopy of Firenze. In particular, I will focus on the studies of the radiation produced in the extreme ultraviolet by the process of high-order harmonic generation, and on the white-light supercontinuum obtained when very intense laser pulses impinge on transparent materials.

I will show how, using very simple experimental ideas to investigate the coherence properties of these two different kinds of ”extreme” light sources, we were able to obtain very interesting and unexpected results. I will also try to show how, by looking at the outcome of such experiments from a wider perspective, it was possible to find new and surprisingly fruitful results in fields that could initially appear as almost completely uncorrelated, like strong-field atomic physics and high-precision optical metrology.

2 Spectroscopy with sequences of pulses

The driving idea for our experiments was the possibility of using ultrashort light pulses of various kinds to perform high-resolution spectroscopy. This is one of the experimentalist's dream and would allow to exploit the extremely high peak intensities characteristic of the short pulses to excite multiphoton processes, or to drive highly nonlinear phenomena (and generate new wavelengths, for example), while maintaining the high resolution characteristic of CW sources to investigate very narrow spectral structures of atoms or molecules.

Of course, the two conditions of ultrashort pulse duration and high spectral resolution normally appear in striking contrast, since short pulses invariantly correspond to broad spectral bandwidths that limit the frequency resolution to the inverse of the pulse duration. However, if pairs of time-delayed and phase-locked pulses (like those generated by splitting a single laser pulse by means of a Michelson interferometer) are used, a simple Fourier transformation shows that the corresponding spectrum maintains the broad bell-shaped envelope, but also acquires a sinusoidal modulation with a spectral period given by the inverse of the temporal separation between the two pulses. It is this fringe period that now sets the instrumental resolution and allows one, in principle, to investigate very fine spectral features if long time delays are available.

We demonstrated that this was indeed the case in 1996, during my first collaboration with Ted Hänsch, when we showed that it was possible to measure line splittings (the hyperfine separation of the $8S_{\frac{1}{2}}$ state in cesium in that case) in a two-photon transition with a spectral resolution much better than that given by the single-pulse spectral width [1].

The idea of using a pair of phase-locked pulses in order to achieve a better spectral resolution can also be extended by the use of longer sequences of equally time-delayed and phase-locked pulses. The spectrum that one obtains in this case still presents the broad bandwidth connected to the short pulse duration, but is now modulated in a sharper and sharper fashion as longer pulse sequences are used (see Fig.1). In the ideal limit of an infinite train of phase-locked and equally spaced pulses, the resulting spectrum essentially consists of a "comb" of infinitely sharp lines, equally separated by a frequency spacing corresponding to the inverse of the interpulse period.

The potential advantages of such a peculiar spectral distribution are evident: if such a comb is realized in practice, it can be used as a precise ruler to measure unknown frequency intervals in a relatively simple way. By locking two laser lines to two different "teeth" of the comb, and by counting the integer number of interposed teeth, one can immediately obtain the unknown frequency gap if the separation between the teeth is well known.

A mode-locked laser is a natural way for generating such an ideally infinite sequence of time-delayed pulses with a well defined phase relationship [2]. Its spectrum (given by the set of equally-spaced longitudinal modes of the cavity) is a broad comb of frequencies with a mode separation equal to the measurable and controllable pulse repetition rate. The largest frequency gap that can be bridged with such a comb is determined by the inverse of the

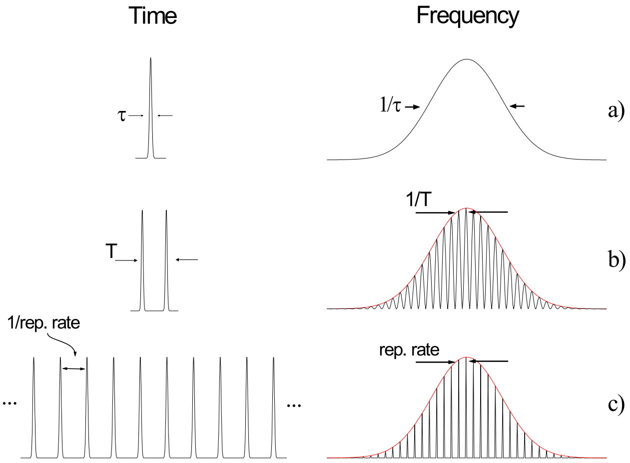

Fig. 1. A single laser pulse has a frequency bandwidth which scales with the inverse of its duration τ. If one uses a pair of phase-locked pulses delayed by a time T, the resulting spectrum maintains the broad envelope of width $1/\tau$ but with a sinusoidal modulation of period $1/T$. This width sets the new instrumental resolution. If one uses an infinite sequence of pulses locked in phase and equally delayed in time of a time interval T, the spectrum breaks up in a "comb" of very narrow lines (the "teeth") equally spaced by a frequency interval $1/T$.

pulse duration, but it can be widely extended if nonlinear interactions are used to broaden the spectrum.

These ideas are some of the best examples of the importance of having such a clear and wide perspective on physics as to imagine useful connections between normally separated worlds. As a matter of fact, people dealing with ultrafast lasers usually think of their applications only in the temporal domain, while spectroscopists generally believe that ultrastable lasers are necessary to do their job and only speak in terms of frequency. It is instead easy (once someone has shown you the way) to get the best of the two worlds and to do so without a growth in complexity, but rather with an enormous increase in the range of possible applications.

In any case, one of the essential requisites for these novel spectroscopic techniques to work, is that the phase coherence between the pulses in the sequence is accurately preserved. The experiments described below were mainly aimed to check this coherence preservation in particular highly-nonlinear processes, but their results went far beyond the initial scope.

3 High-Order Harmonics

At the time of the two-pulse experiments on cesium, we had already developed at LENS a reliable source for the generation of high-order harmonic pulses and we were still mainly characterizing this radiation while trying to think of clever ways to use it for spectroscopy in the vacuum and extreme ultraviolet (VUV and XUV) [3].

Pulses with frequencies which are odd-order harmonics of the fundamental laser frequency are generated by the interaction of short and intense laser pulses with the atoms of a supersonic jet. Depending on the wavelength, duration and peak intensity of the pulses, and on the ionization potential of the atoms, very high orders can be efficiently generated at wavelengths down to the XUV or to the soft x-ray regions.

Though harmonic sources are extremely appealing due to the lack of other easily accessible alternatives in these spectral regions, it looked like no true spectroscopy would have ever been done with them, due to the extremely broad bandwidth associated with the short duration of the harmonic pulses. In fact, even if some low-order harmonics can be generated with pump pulses in the picosecond range, allowing one to keep an acceptable spectral resolution for selected applications, higher-order harmonics can only be created at intensities above 10^{13} W/cm^2 by ultrashort laser pulses; and a 100-fs pulse is already characterized by a spectral width in the THz range.

We immediately thought of overcoming this limit with the application of the two-pulse technique to the harmonic radiation, by splitting and delaying the XUV pulses by means of a Michelson interferometer before sending them to the samples under study. Unfortunately, the use of this technique with harmonic pulses is far from straightforward, mainly because good interferometers cannot be built to work in the XUV due to the lack of suitable optics.

A possible solution to the problem was to invert the two steps of harmonic generation and pulse splitting, by moving the interferometer in the path of the laser beam, in order to create two phase-locked and time-delayed pump pulses that would have generated equally phase-related XUV pulses. The question at this point was about the preservation of the phase lock in the generated pulses: if the process of harmonic generation was an incoherent one, no phase relationship would have been preserved between the XUV pulses and the whole scheme for spectroscopy with harmonics would have been useless.

A simple way to test the mutual phase coherence between the harmonic pulses was to generate them in two separate spatial regions and look for interference fringes in the far field: the existence of an interference would have shown that the two secondary sources have preserved a memory of the phases of their parent pulses.

We first performed a preliminary experiment with the third harmonic generated in air and, though it was not possible to directly extrapolate the results to higher orders, it gave very interesting and encouraging indications

[4]. At the same time, Ted Hänsch discussed these ideas with Anne L'Huillier, one of the major world experts of high-order harmonics, and, though she initially suspected that the generation process would have completely messed up the phases of the pulses destroying any interference, we agreed to do a joint experiment to test these ideas.

The first experiment was carried out in Lund, where harmonics were generated by focusing 30 ps laser pulses into an argon gas jet. After the focusing lens, the pulses coming from the laser where splitted and given a slight fixed displacement (both in space and time) thanks to the walk-off in a birefringent plate. A common polarization component was selected before entering the interaction region. Harmonics were then generated in two different positions in the gas jet and we looked for interference fringes in the far field after the spectral dispersion operated by a grating. We unexpectedly observed nice, stable and highly-contrasted fringes, indicating that the generation process was not as phase-destructive as we initially thought [5], and demonstrating that harmonic generation was a suitable source for the high-resolution spectroscopic technique we had in mind.

The second, more accurate, experiment was done at LENS. Here we used much shorter (100 fs) laser pulses and we splitted and delayed the pulses by means of a Michelson interferometer, so that their temporal and spatial separation in the focus could be carefully adjusted and controlled.

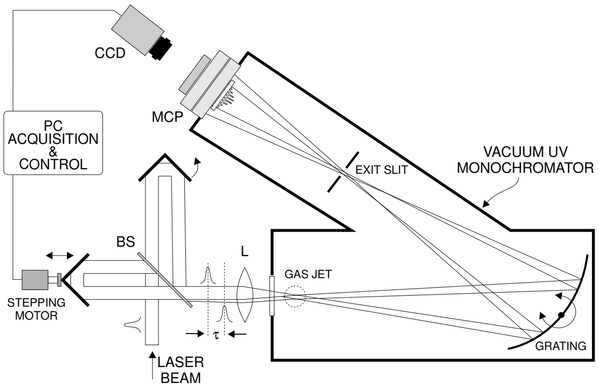

Fig. 2. Scheme of the experimental setup used at LENS for the observation of the interference fringes and for the measurement of the temporal coherence of high-order harmonic pulses. The pump laser pulses are splitted and delayed, and their output directions are slightly tilted by means of a Michelson interferometer, so that the two beams are focused in different positions of the gas jet and generate harmonics independently. We looked for interference fringes on a Micro Channel Plate detector placed after the exit slit of a vacuum monochromator.

Again, we saw very clear interference fringes on the Micro Channel Plate detector placed beyond the exit slit of our vacuum monochromator, and we were also able to measure the temporal coherence of the harmonics by observing the decay of the fringe visibility as a function of the delay [6].

We demonstrated that one can generate almost transform-limited XUV pulses with coherence times of the order of the expected duration of the harmonics themselves (about 40 fs), showing that not only the phase is not scrambled in the process, but also that a negligible frequency chirp is imparted to the secondary pulses.

While doing this we also discovered something that was quite a mystery at the beginning: the presence of two clearly distinct spatial regions in the pattern of harmonic emission, with drastically different coherence properties (see Fig.3). An inner region with the long coherence times described above, and a diffuse outer halo, containing more than half of the total emitted flux, with an extremely short temporal coherence, of the order of few femtoseconds.

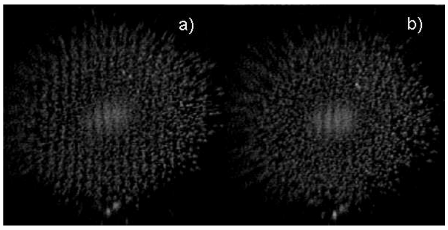

Fig. 3. Snapshots of the interference fringes produced by the 15th harmonic generated in argon. In a), taken at a delay of 0 fs, fringes appear all over the image with a good visibility, while they disappear in the outer region of b), taken at a delay of 15 fs. The diffuse halo surrounding the central bright spot has a much shorter coherence time than the inner region.

We unexpectedly found that this was one of the most direct proofs of the validity of current theoretical models used to describe the microscopic physical mechanisms involved in the process of harmonic generation.

According to the standard picture of the high-order harmonic generation process [7], every half optical cycle of the laser pulse, electrons undergo tunnel ionization through the potential barrier formed by the atomic potential and by the electric field potential of the laser; after being accelerated in the ionization continuum by the field, they may come back to the ion core and finally recombine to emit harmonic photons that release the accumulated kinetic and ionization energy. Single-atom models also predict that harmonics are emitted with an intensity-dependent phase, proportional to the amount of time spent in the continuum by the generating electrons.

Simple calculations show that the highest harmonic orders (in the so-called *cutoff*) can only derive from electrons which have been released at a well defined time in each half optical cycle of the laser. On the other hand, it is equally easy to see that lower-order harmonics (in the so-called *plateau*) essentially come from two different classes of electrons which are emitted at different moments, spend different amounts of time in the continuum following different trajectories, but nevertheless come back to the ion with the same correct kinetic energy to generate that given harmonic (see Fig.4).

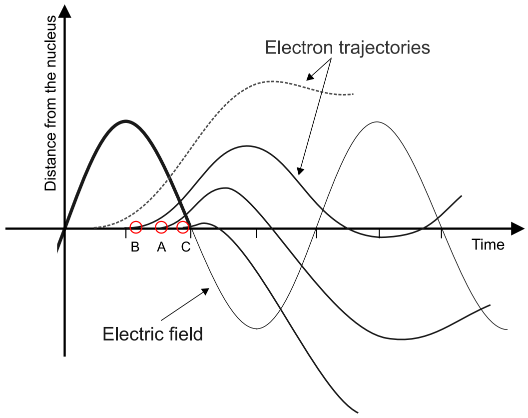

Fig. 4. Schematic representation of the possible classic electronic trajectories leading to the emission of harmonic photons. Only half optical cycle of the laser is considered here since everything repeats with this periodicity. Electrons which are ionized when the field amplitude grows are driven away from the ion and never recombine to generate harmonics (dashed curve). Electrons escaping with a decreasing field will leave the ion, oscillate in the continuum and come back with some supplementary kinetic energy that can be converted into photon energy (solid curves). The maximum return energy (corresponding to the highest harmonic orders generated) is carried by electrons ionized in A. A given lower harmonic can be generated by electrons released either in B or in C, since both trajectories correspond to the same final return velocity (same angle of intersection with the horizontal axis). Electrons "born" in B suffer a stronger intensity-dependent phase modulation since they spend a much longer time in the continuum.

For these harmonics, the "short" trajectory imposes a phase that does not vary much with the laser intensity, whereas the phase corresponding to the "long" trajectory varies rapidly with the laser intensity. Such a "long" electronic trajectory gives rise to strongly divergent angular emission because the rapid spatial variation of the phase with the focused laser intensity leads to a strong curvature of the phase front. This radiation also has a very short coherence time, since the harmonic pulse is strongly chirped due to the rapid temporal variation of the phase during the pulse. In contrast, the phase variation corresponding to the "short" trajectory is much less important. The emitted radiation has a long coherence time and is much more collimated (for a more detailed discussion see ref.[8]).

Our simple experiment allowed us to directly observe these effects for the first time. It was somehow amazing that, just from the observation of the appearance and behavior of interference fringes, we were able to gain such a deep insight on the inner dynamics of atoms in the presence of strong laser fields.

Moreover, some of the most interesting latest developments in the field of high-order harmonics have been a more or less direct consequence of these results: harmonic sources have been used for interferometric studies of metals and plasmas in the XUV [9], and the possibility of using pairs of harmonic pulses for high resolution spectroscopy has also been recently demonstrated [10]. Finally, our proof of the role of the different electron trajectories in the process of generation is giving a substantial push in the race towards the attosecond barrier.

4 White Light

In 1997, while we were still performing the experiments on the coherence of high-order harmonics, we decided to use our simple experimental apparatus to test the mutual phase coherence of the light pulses generated in a different kind of extremely nonlinear process. We just replaced the system for harmonic generation with a plain plate of calcium fluoride (CaF_2) to generate pulses of white light.

The process of white-light continuum generation provides a simple and efficient way to achieve an extreme spectral broadening, by focusing intense enough laser pulses into transparent materials [11,12]. The dominant process leading to spectral super-broadening is the self-phase-modulation of the pulse due to an intensity dependent refractive index of the medium, but a number of other linear and nonlinear effects play a role as well, including self-focusing, parametric four-photon mixing, stimulated Raman and Brillouin scattering and shock-wave formation. The generation of the continuum is then the result of a very complex interplay between competing processes and the 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 might loose any precise phase relationship in the generation process.

Again, like in the case of harmonics, we were rather pessimist about the possibility of a residual phase coherence between the resulting pulses but, since we had been lucky the first time, and since it was a very simple experiment to perform with our existing apparatus, we just decided to give it a try.

A 2 mm thick CaF_2 plate was placed in the focal plane of a lens after the Michelson interferometer as shown in Fig.5, so that the two phase-locked laser pulses could independently produce two white light pulses. After the interaction zone and after the two diverging continua had propagated in air for some distance, they were finally overlapped on a screen where we hoped to observe interference fringes.

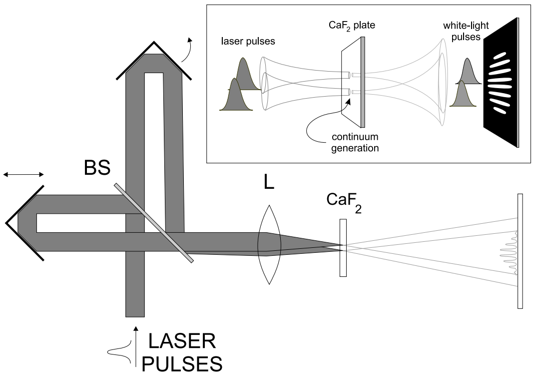

Fig. 5. Scheme of the experiment for the observation of white-light interference fringes. The two pump pulses are generated by the same laser system and by the same Michelson interferometer described above. Two independent supercontinuum pulses are generated in the material and then propagate and overlap on a screen.

Our results were once again unexpected and intriguing: when the two pump pulses were properly balanced in intensity and adjusted for zero relative delay, the two white-light continua that they generated separately showed the surprisingly clear and stable white interference fringes shown in Fig.6, indicating that we were dealing with highly phase-correlated secondary sources [13].

It is instructive 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 experiments on harmonics and white light, on the contrary, the interference fringes appeared because of the spatio-temporal superposition of two secondary light pulses independently generated in two separate positions of the medium. For the complex and apparently unpredictable characteristics of the generation processes at play, one could expect such pulses to be highly uncorrelated.

In general, in order not to damage the crystals by destructive optical breakdown and to avoid multiple filamentation in the medium, we limited the energy of each 100-fs pulse coming from the interferometer to less than 3 μJ. Under such conditions, a single filament was generally created by each focused beam in the transparent material, with an estimated peak intensity of the order of 10^{12-13} W/cm^2. Nonetheless, we observed that even in the case of multiple filaments and depending on their number, more or less complicated interference structures appeared, indicating that the mutual coherence among the resulting continua was conserved.

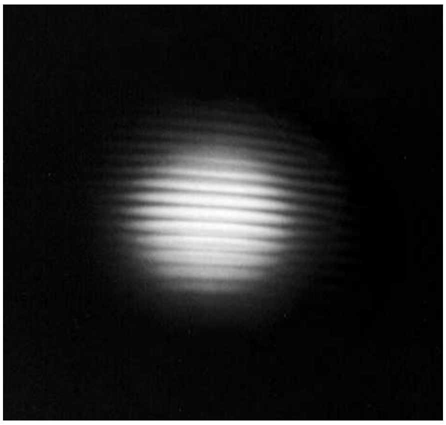

Fig. 6. Snapshot of the observed interference pattern. This picture was taken with an exposure time of about 2 seconds and with a laser repetition rate of 1 kHz. The very good visibility of the fringes indicates that the phase lock is not only preserved on a shot-to-shot basis, but that a constant phase relationship is maintained over at least thousands of shots.

After the first appearance of the white-light fringes, we also tried to generate similar fringe patterns from different materials: quartz plates and plain glass microscope slides proved effective to produce a stable interference, as well as water cells and Plexiglas plates. In general, any transparent material that we could find in the laboratory was able to produce nicely interfering white light at various degrees of efficiency.

We obtained an even more visual demonstration of the effective phase-lock over the whole visible spectrum by recording the spectrally dispersed interference pattern. We used a planar diffraction grating and a cylindrical lens, 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 was possible to produce stable fringe patterns of high contrast, as shown in Fig.7. This was another clear indication that the two continua were mutually phase-coherent over the entire visible spectrum.

Though the experiment was of an extreme simplicity (it took just one day to see the first white fringes), it nonetheless bore very important consequences. After the first surprise at the look of the nice interference patterns, it soon became evident that, by demonstrating the phase preservation in the process of supercontinuum generation, we had also shown that it was possible to generate a sequence of phase-locked white-light pulses from a sequence of phase-locked pump pulses. In other words, we had shown that it was in principle possible to build a white-light frequency comb.

The fantastic possibilities of such a device were already clear to Ted (not quite so clear to me at the beginning!): one could build an extremely "long"

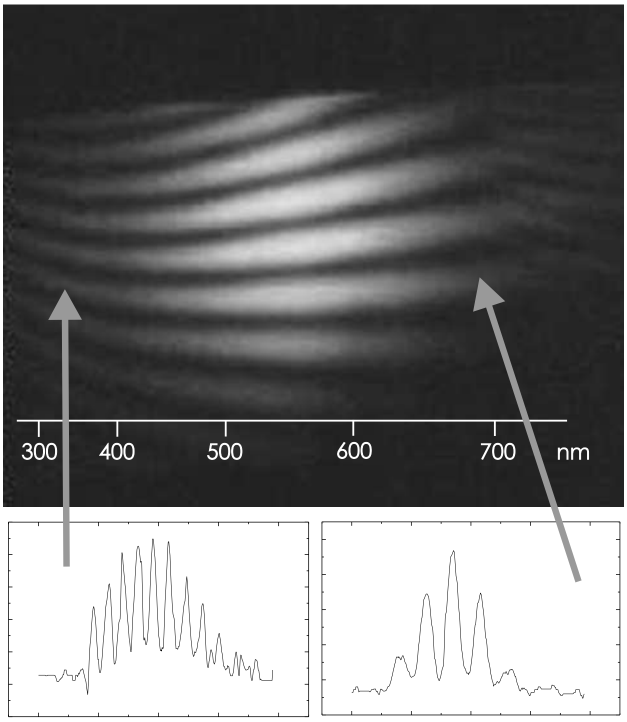

Fig. 7. Spectrally dispersed white-light fringes (note that in the original picture the color of the fringes changes from violet to red when moving from the left to the right side of the screen). Clear and well defined fringes indicate that a stable phase relationship is conserved across all the generated 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 larger than that of the fringes in the left graph.

and precise ruler to measure optical frequencies throughout the whole visible range in a single step, doing away with clumsy laser frequency chains, divider stages, and so on.

The ideal path to the generation of a white-light comb involved the use of a train of phase-locked infrared pulses from a mode-locked laser to produce a train of phase-locked, white-light pulses after the interaction with a medium. Only two main obstacles could hinder the practical realization of such a wide comb: the first one was the possibility of phase scrambling between successive pulses in the output train, but, with our experiment, we had demonstrated that this was not the case. The second one was of a more technical nature, and depended on the limited pulse energy of mode-locked lasers. At the nanoJoule-level energy characteristic of these high-repetition-rate systems, it was absolutely impossible to generate a supercontinuum and, unfortunately, no solution to this problem was available at that time. So, although these experiments had been performed in 1997, we kept all the nice and colorful pictures we had recorded aside for quite a long time because, without the possibility of such an application in sight, we just did not know what to do with them, apart from showing them around among the general amazement.

Finally, in 1999, a new kind of optical fibers came out [14] with new, exceptional properties: these "photonic crystal fibers" or "holey fibers" are essentially constituted by a very small silica core surrounded by a regular structure of "holes". They have the very interesting characteristic of supporting the propagation of highly spatio-temporally confined visible pulses over long distances. This allows for high nonlinearities to build up along the fiber and can give rise to the generation of a supercontinuum already from nanoJoule-level pump pulses.

The results of our experiment and the introduction of these new fibers boosted the research toward the realization of white-light combs. Such devices are now a reality and are revolutionizing the whole field of optical metrology and spectroscopy. Femtosecond frequency combs are now replacing old-style frequency chains wherever a precise measurement of an optical frequency is required. With the realization of combs so wide as to extend over more than one optical octave, this technique now constitutes a self-referencing method allowing the measurement of absolute optical frequencies with extremely high accuracy in a single step from the frequency standard [15].

5 Conclusions

To conclude, I hope I was able to give an impression of the kind of nice work that can be produced from the interaction with such a source of an amazing number of clever ideas as Ted Hänsch. Ranging from the apparently simple ones (like the suggestion he once gave us on how to time-share the pulses from our laser among different simultaneous experiments: why had we never thought about it before ?), to those that have really revolutionized the history of laser physics, there is always a good and unexpected idea waiting to pop out, thanks to his immense enthusiasm and curiosity towards science. It has been, and I hope it will be for a long time to come, a great pleasure to contribute to the realization of some of these ideas.

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