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PICOSECOND PULSES FROM UV TO IR USING A SPECTRO-TEMPORAL SELECTION DYE LASER

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1. METHODS OF CONTINUUM GENERATION

Ultrafast spectroscopy, in the femto and picosecond domain, requires single pulses at different wavelengths and/or a spectral continuum of corresponding duration. Basically the same simple technique is used to generate the so-called "super-continuum" from ps or fs pulses though the non-linear phenomena involved in the generation of new frequencies are different [1,2]. The laser beams are focused in a refractive medium, such as high pressure gas [3], liquid, glass or any non absorbing material. The spectrum of the output light is found to be broad, more or less continuous when averaged over several shots. The spectrum depends in shape and intensity on the input pulse duration, intensity and waveform. Theoretical description of the spectral broadening [4] is still progressing [1,5]. With 50-100 femtosecond pulses amplified to high energy (≈ 1 mJ), the continuum, essentially due to self-phase modulation [2], is easily produced by focusing in a cell or in a thin liquid jet, but the production of powerful femtosecond pulses is much less straightforward, requiring expensive, delicate and bulky equipment, with two pump lasers (one CW, one pulsed), several dye laser systems (a colliding pulse modelocked ring laser, several amplifiers) and electronic synchronization between the different repetition rates [6,7]. On the other hand, high energy picosecond laser pulses, typically 10 mJ and 20-40 ps, are more easily produced with one pulsed laser but, when focused in a long (~ 10 cm) cell, they can generate only a strongly structured continuum, suggested to be due to a mixture of several non-linear processes [8,10] such as stimulated Raman, four-photon parametric interaction, self-phase modulation [8,10] etc... Self focusing and moving focus effects contribute to the strong spectral instability observed which originates in the low transverse beam quality of standard solid state lasers. Such a picosecond continuum is routinely used for picosecond spectroscopy [11,12] but requires sophisticated detection electronics with a reference measurement for each pulse.

Simple ways of generating a broad and stable continuum will perhaps emerge from the several recently proposed new methods of producing picosecond and sub-picosecond pulses. These methods have in common that they use short dye cavities and start from non-mode-locked laser pulses, which is a noticeable simplification and widens the choice of possible pump lasers. They use the processes of cavity transients [13-16], cavity quenching [17], distributed feedback [18-20] or spectro-temporal selection (STS) [21-24]. They can be used in a cascade configuration [15,19,21] with successive stages of pulse shortening, separated by amplifiers pumped by a fraction of the same pump pulse. They can be combined in a great variety of ways. The STS method has the largest efficiency in pulse reduction; a time reduction of four orders of magnitude with a two-stage scheme was demonstrated [22]. Two methods were recently reported to be able to produce subpicosecond pulses : one uses a three-stage reduction process [20], the other uses subpicosecond

modulations from mode beating [23]. They are of interest for the generation of a very broad band continuum. We report below the production of powerful ps or subpicosecond pulses, with or without femtosecond modulation and their use for continuum generation from UV to IR.

2. PRODUCTION OF PICOSECOND PULSES, STARTING FROM AN STS DYE LASER.

2.1. General principle

The two-stage scheme used here can reduce an initial ~ 10 ns pump pulse by two orders of magnitude at each stage, i.e. down to ps or less. After each stage the shortened pulse is amplified in saturated multipass amplifiers pumped with the 10 ns pulse. In the first stage the spectro-temporal selection (STS) method [22] is applied to a short and low-Q cavity to obtain a ~ 150 ps pulse by selecting wavelengths in the blue wing of its sweeping broadband emission. In the second stage this 150 ps pulse pumps a microcavity filled with another dye solution. The emission of this second oscillator can be used in two different ways. First, one can again use the STS method to isolate the emission of a single mode in the blue wing of the multimode emission, which produces a nearly transformed limited pulse of 1.5 or 4 ps with cavity lengths of 100 μm or 200 μm [22]. The second way is to amplify the total (multimode) emission, producing

Fig.1 Two-stage spectro-temporal selection picosecond dye laser and amplifiers. O_1 and O_2 : sweeping oscillators, FR : Faraday isolator A, A' : amplifier cell ; AS : saturable absorber ; G_1 , G_2 = gratings

pulses temporally modulated at the cavity roundtrip time owing to mode beats. For example, for a 30 μm cavity a two mode emission can be obtained giving pulses modulated into a 1 ps train of femtosecond pulses. After amplification to a few tenths of mJ these ultrashort pulses can generate supercontinuum from UV to IR when focused in any transparent medium.

2.2. First reduction stage : from 10 ns to 100 ps

About 300 μJ of a 532 nm 10 ns pulse from a standard Q-switched 10 Hz Nd:YAG laser, pumps through a cylindrical lens, a 1 cm cell with Rhodamine 6G in ethanol, $3 \cdot 10^{-4}$ M/l, which constitutes a short and low-Q ($R \sim 0.04$) sweeping oscillator [21] (fig.1), operated about 4 to 6 times above threshold. The frequency sweeping laser emission is collimated with a lens and filtered on a grating tuned to select a 0.3 nm line at 560 nm in the blue wing. The output is typically a 100-150 ps pulse which is amplified by a factor 10^4 to 10 μJ in multipass amplifiers. Its shot to shot stability in duration and intensity very much depends on the stability of the 532 nm pump pulse substructure. The use of highly saturated multipass amplifiers reduces the intensity fluctuations to less than 30 %. A typical shape of this pulse is shown in Fig.2a.

Fig.2 Single sweep streak camera trace of the dye laser pulses
a : First stage pulse
b : Second stage pulse, delayed.
(Camera CS-1 from Société ARP)

2.3. Second reduction stage : from 100ps to 1ps.

The 100ps pulses at 560 nm are focused with a 5 cm lens on a multimode low-Q microcavity (O_2 in Fig.1, length between 30 and 200 μm) filled with Rhodamine 640 in ethanol or methanol with a concentration between $5 \cdot 10^{-2}$ and $5 \cdot 10^{-3}$ M/l according to the cavity length. The mirror reflectivities are low: $R_1 = R_2 = 0.1$, or lower. One of the plates - or both - can have no coating at all. The Fresnel reflection coefficient, $R \sim 10^{-3}$, between the silica plate and the alcohol appears to be sufficient to

Fig.3 Time integrated spectrum of a 100 μm cavity dye oscillator

Mode spacing : 1.3 nm

initiate the laser emission, well characterized by the longitudinal mode pattern of the time integrated spectrum (Fig.3). Single pass amplification of more than 10^3 means that about 30 % of the dye molecules are in the excited state, with an average stimulated emission cross section of $\sim 2 \cdot 10^{-16}$ cm^2 , and an average absorption cross section, at the same frequency, one order of magnitude smaller. The output pulse shape does not follow the pumping waveform, as expected in the case of relaxation oscillations [13]; a very strong initial spike occurs quasi-simultaneously for all modes [22]. The subsequent spikes are much weaker and exhibit spectro-temporal evolution [22,24]. However it should be noted that other damping processes, such as molecular orientational relaxation, were not taken into account in the analysis. They may enhance the relative magnitude of the first spike.

The output pulse from the multimode microcavity can be used in two ways:

a) A second spectro-temporal selection in the blue wing can give a single spike pulse for all shots, with a FWHM duration of less than 2 roundtrip times, i.e. 4 ps with a 200 μm cavity, 1.5 ps with a 100 μm cavity. These pulses can be amplified as shown in Fig.1. In this case the filter used must not lengthen the pulse (high reflectivity Fabry-Perot and grazing gratings are excluded). In these experiments we used a 7 μm thick Fabry-Perot with $R=0.95$ (cavity lifetime 0.4 ps), or a single pass grating with a spot diameter less than 1mm (transverse delay < 1ps).

b) For a given distance between the pump focusing lens and the microcavity, and for a pumping of about 3 to 4 times that of the threshold, an output laser pulse with only one spike could be obtained with no spectral filtering for most of the 10 Hz pump shots, as was shown by permanent sampling, at 1 Hz, of the pulse shape with a streak-camera (fig.2b). The number of strongly excited modes depends mainly on the mode spacing and on the gain spectrum. With a ≈ 30 μm long cavity the mode spacing is measured to be 3.5 nm, and with rhodamine 640 in methanol at $c = 3 \cdot 10^{-2}$ M/l, only one (or two) modes have a strong intensity, with two (or one) additional weak modes. The overall pulse duration was found by intensity autocorrelation to be of the order of 1 ps or less ; assuming a sech^2 envelope shape, a 600 fs pulse duration was found for a 3 mode emission. The time modulation corresponding to the modulated spectrum is obvious in the total autocorrelation trace in fig.4. A more detailed analysis is possible by cross-correlation as shown in §2.4.

Fig. 4 Total auto-correlation trace for the modulated pulse from a 30 μm cavity dye oscillator.

2.4. Analysis of the multimode pulse from a 30 μm cavity

The output of the 30 μm multimode microcavity is modulated at 2.9 THz which corresponds to the cavity roundtrip time $T = 340\text{fs}$. Phase information can be obtained by recording the cross-correlation trace with the standard non-collinear autocorrelation device with addition of

Fig. 5 Background free auto-correlation scheme with addition of a grating to separate the UV spectral components.
F : UV filter .

Fig. 6 Cross correlation trace at the sum-frequency (a) and auto-correlation trace at a doubled frequency (b) for a 3 mode pulse (30 μm cavity).

a grating to analyse the UV output (fig. 5). When the 1mm thick KDP crystal is adjusted for non collinear frequency doubling, the UV light comprises three spectral lines at $2\omega_1$, $2\omega_2$ and $\omega_1 + \omega_2$ for a two mode pulse at ω_1 and ω_2 . A 3000 g/mm grating and a slit are used to record separately the sum-frequency intensity, or the doubled frequency intensity, as a function of the correlation delay τ . The measured cross-correlation trace in the case of a 3 mode emission with only one intense mode ω_2 , is given in fig. 6a and shows a complete modulation for $\omega_1 + \omega_2$ while the doubled frequencies $2\omega_2$ give as expected little modulation. (A small modulation can occur since $\omega_1 + \omega_3 = 2\omega_2$). From the intensity second order correlation function a maximum occurs when τ is incremented by a cavity roundtrip time T . We have:

$$I_{\omega_1 + \omega_2}(\tau) = \eta^2 G^{(2)}(\tau) \cos^2(\pi\tau/T)$$

assuming two modes of equal amplitude and in phase; η is the efficiency of sum-frequency in KDP; and $T = 2\pi/(\omega_1 - \omega_2)$. From fig. 6a each peak is estimated to have a duration between 100 and 150 fs. No significant phase shift can be noted.

3. CONTINUUM GENERATION

By focusing the fs-modulated ps pulse in a 1cm cell of water a strong continuum is generated as shown by the white light emerging from the cell observed with no filter. This light was collimated with an achromat lens and its spectrum recorded with a Jarrell Ash polychromator

equipped with an SSR OMA. Fig.7 shows the spectrum from 350 to 1100 nm obtained with a 2 ps modulated pulse from a 100 μm cell. The spectrum averaged on 300 laser shots was recorded by sections of 300 nm. Appropriate Wratten filters and dye solutions were used to attenuate the

Fig.7 Spectrum of the continuum obtained with a modulated 2ps pulse.

central part of the continuum and the laser line. The transmitted laser line through the 1 cm cell remains strong for pulses longer than 1ps. Comparison between the continuum generated by a single mode pulse (no mode beating) and by a two-mode pulse with same total energy (THz mode beating) showed that the intensity of the continuum is 2 to 3 times stronger with the modulated pulse, and does not exhibit the additional broadened Raman-like bands obtained with the non modulated pulse. Fig.8 shows the central part of the continuum attenuated by a DCI2 solution which reduces the laser intensity by a factor ≈ 8 . The main dip in the blue wing is due to DCI2 absorption secondary maximum.

Preliminary measurements of the duration of the continuum pulse by cross correlation with the laser pulse showed no clear lengthening of the pulse duration. Measurement of the continuum pulse with the streak camera showed no delay between the blue and red part of the spectrum within the camera resolution of ~ 5 ps. When the laser pulse focused in water was deliberately made (by over pumping the microcavity) to consist in a succession of several irregular spikes of similar intensity and

Fig.8 Effect of pulse modulation on the central part of the continuum spectrum (normalized to the transmitted laser intensity).
a) single mode pulse at 603 nm.
b) modulated two mode pulse at 603 and 607 nm.

Fig.9 Single continuum spike generated by a two-spike laser pulse (delayed) observed with a streak-camera ARP.

separated by 10 to 100 ps, it was noted that only the first spike generates a continuum pulse. This effect, not yet explained, is of advantage to produce clean single ps pulses from UV to IR.

4. FREQUENCY CONVERSION BY TRAVELLING WAVE AMPLIFICATION IN DYES

Direct conversion of the powerful ps laser pulses at 603 nm into red or near IR pulses was also obtained by focusing the laser beam into a lcm cell containing a dye solution (the concentration can be adjusted so that the pump laser beam is completely absorbed). With the cell length being much longer than the length of the pulses, travelling wave amplification of the fluorescence occurs in the direction of the incident laser beam. With this very simple technique, ps pulses were obtained at a definite fluorescence wavelength of the dye; for example at 660, 720 and 850 nm with DQTCI, DOTC and IR 140 dye solutions in alcohol. The spectral width of the output, of the order of 9 nm, can vary by a factor 2 as a function of the dye concentration and on the incident photon flux. Observation of output pulse durations with the streak camera showed also that when the incident laser pulse is spiked, only the first spike generates the Stokes shifted pulse. Several percent of the initial laser pulse were converted into the downshifted frequency.

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