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Dispersion Effects on Femtosecond Laser Pulses

C.H. Brito Cruz
Universidade Estadual de Campinas (UNICAMP)
Instituto de Fisica
Depart. de Eletronica Quantica
Campinas, Brazil

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Carlos H. de Brito Cruz
Departamento de Eletrônica Quântica
Instituto de Física, UNICAMP
13081, Campinas, SP, BRASIL

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1 Introduction

In these notes we discuss the effects of dispersion on the duration and shape of femtosecond duration laser pulses. As the duration of the available laser pulses became shorter than a few hundreds of femtoseconds, the corresponding bandwidth became so large such that dispersive effects, associated for example with the propagation through material media, became important to the extent that severe pulse distortion can happen upon propagation. Some clever devices have been devised, with which it is possible to compensate for the distortion caused by dispersion, so that today pulses as short as 6 femtoseconds can be generated[1].

Consider for example a pulse with a duration of 6 femtoseconds and a central wavelength of 620 nm. With this duration the pulse envelope covers only three optical cycles of the carrier wave. The associated bandwidth is larger than 65 nm, and spans most of the visible range. Such a pulse will have its duration doubled upon propagation through 1 mm of quartz. In order to perform experiments using these pulses it is necessary to propagate them through lenses, mirrors and other optical elements so that it is essential to know what kind of distortion shoud be expected and, better still,

[1] R.L. Fork, C.H. Brito Cruz, P.C. Becker and C.V. Shank, Opt. Lett. 12, 483 (1987)

how to control and compensate it. In pulse compression systems it is also necessary to use adequate compensation for distortion due to dispersion, if the best short pulse for a given system is desired[2].

2 Pulse propagation and dispersion

To consider the effects of dispersion on an ultrashort laser pulse it is more convenient to represent the pulse in the frequency domain. Let us consider a pulse described in the time domain as

$$e(t) = e_0(t) exp\{j[\omega_0 t - \phi(t)]\}$$

with Fourier transform given as

$$E(\omega) = E_0(\omega) exp[j\Phi_0(\omega)].$$

Upon propagation through a dispersive medium the pulse will accumulate a phase shift, which is frequency dependent, given as $\Phi(\omega)$. This phase shift contains the effect of the dispersive pproperties of the medium. For example, in the case of propagation through a length of quartz, with a refractive index $n_q(\omega)$, the accumeultaed phase shift will be simply $\Phi(\omega) = \frac{\omega}{c} \times n_q(\omega)$. According to the particular dependence of $\Phi(\omega)$ on the angular frequency, different types of pulse distortion (or even distortion correction) can occur.

2.1 Phase distortion

The accumulated phase shift can be expanded as a Taylor series

$$\begin{aligned}\Phi(\omega) \;=\; & \Phi(\omega_0) + \frac{d\Phi(\omega)}{d\omega}(\omega_0) \times (\omega - \omega_0) + \\ & \frac{1}{2}\frac{d^2\Phi(\omega)}{d\omega^2}(\omega_0) \times (\omega - \omega_0)^2 + \frac{1}{6}\frac{d^3\Phi(\omega)}{d\omega^3}(\omega_0) \times (\omega - \omega_0)^3 + \\ & \frac{1}{24}\frac{d^4\Phi(\omega)}{d\omega^4}(\omega_0) \times (\omega - \omega_0)^4 + +\ldots\ldots \qquad (1)\end{aligned}$$

[2]See notes by A.M. Johnson on 'Pulse compression using optical fibers' on this same Winter College on Ultrafast Phenomena

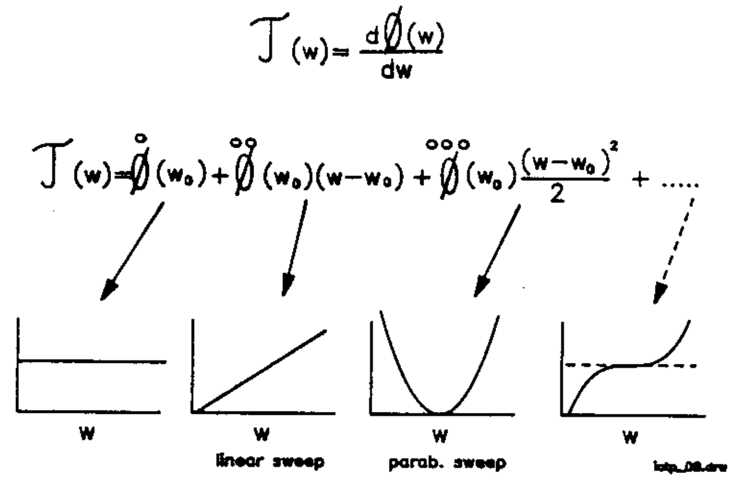

Figure 1: Group delay expansion and related distortion.

Each one of the terms represents a different effect on the pulse shape. The first two terms on the right and side correspond, respectively, to a fixed phase shift and to a delay in the pulse as it propagates. It is easy to show that any linear dependence between the phase and the angular frequency ω does not imply any kind of pulse shape distortion, but only in a shift of the time origin. The higher order terms are the ones that bring distortion into play. It becomes easier to understand their contribution by looking at the corresponding Taylor expansion for the Group Delay, $\tau_d(\omega) = \frac{d\Phi(\omega)}{d\omega}$:

$$\begin{aligned}\tau_d(\omega) \;=\; & \frac{d\Phi(\omega)}{d\omega}(\omega_0) + \frac{d^2\Phi(\omega)}{d\omega^2}(\omega_0) \times (\omega - \omega_0) + \\ & \frac{1}{2}\frac{d^3\Phi(\omega)}{d\omega^3}(\omega_0) \times (\omega - \omega_0)^2 + \frac{1}{6}\frac{d^4\Phi(\omega)}{d\omega^4}(\omega_0) \times (\omega - \omega_0)^3 + \\ & +\ldots\ldots \qquad (2)\end{aligned}$$

The first distortion contribution comes from the second term on the rhs of equation (2). It is related to the second derivative of the phase shift with respect to frequency, and corresponds to a **dispersion of the group delay** around a central value. It causes the group delay to have a linear sweep as a function of frequency. When its value is positive the lower frequencies in the pulse spectrum suffer a delay which is smaller than

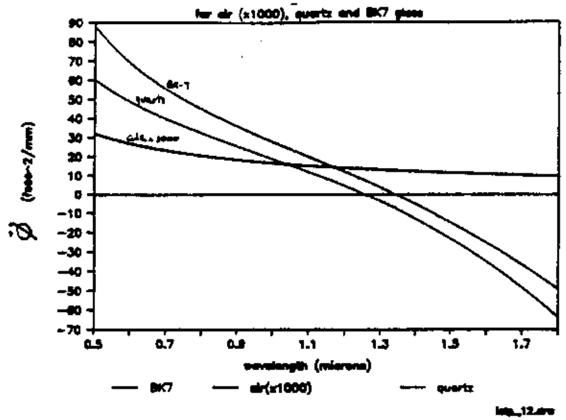

Figure 2: Group delay dispersion for quartz, BK-7 glass and air as a function of the wavelength.

the higher frequencies. The corresponding picture in the time domain is that the lower frequencies of the pulse spectrum are advanced in time with respect to the higher ones, so that the pulse emerges from the dispersive medium with a frequency sweep across its time duration (frequency chirp). For positive values of $\frac{d^2\Phi(\omega)}{d\omega}$ the lower frequencies tend to be on the leading edge of the output pulse, while the higher frequencies tend to be on the trailing edge.

In Figure 2 the value of the Group Delay Dispersion is compared for quartz, BK-7 glass and air. It can be seen that, at the wavelength of 620 nm, 3 meters of propagation through air are equivalent to propagation through 1 mm of BK-7 glass.

From equation (2) it can be seen that the next higher order distortion has a parabolic shape. In this case, for example for a positive value of $\frac{d^3\Phi(\omega)}{d\omega^3}$, both the higher and lower frequencies of the pulse spectrum will be more delayed than the central frequencies. This will give rise to a trailing edge in the time profile of the propagating pulse, with an oscillatory behaviour due to the beating between the high and low frequency components that were delayed.

2.2 Gaussian pulse propagation

For the case of a pulse whose time profile is of a gaussian shape it is possible to calculate in closed form the effect of a liner dispersion of the group delay $(\frac{d^2\Phi(\omega)}{d\omega^2})$. Consider a pulse described as:

$$e(t) = exp[-(2\ln 2)(\frac{t}{T_p})^2] \times exp(j\omega_0 t). \tag{3}$$

Upon propagation trough a system with a quadratic phase distortion given by $(\frac{d^2\Phi(\omega)}{d\omega^2})_s$ the output pulse shape can be easily calculated by taking the Fourier transform of (3), applying the quadratic phase shift

$$phase\,shift = (\frac{d^2\Phi(\omega)}{d\omega^2})_s \times (\omega - \omega_0)^2$$

and inverse Fourier transforming back into the time domain. The result is[3]

$$e_{out}(t) = \frac{1}{[1 + \frac{(\frac{d^2\Phi(\omega)}{d\omega^2})_s^2}{4b^2}]^{\frac{1}{4}}} \times exp\{-\frac{t^2}{4b[1 + \frac{(\frac{d^2\Phi(\omega)}{d\omega^2})_s^2}{4b^2}]} \times exp\{j[\omega_0 t - \phi(t)]\} \tag{4}$$

with:

$$b = \frac{T_0^2}{8\ln 2} \tag{5}$$

$$\phi(t) = -\frac{(\frac{d^2\Phi(\omega)}{d\omega^2})_s t^2}{2(\frac{d^2\Phi(\omega)}{d\omega^2})_s^2 + 8b^2} - 0.5\arctan\frac{(\frac{d^2\Phi(\omega)}{d\omega^2})_s}{2b} \tag{6}$$

2.2.1 Output pulse duration

Equation (4) describes a pulse which still has a gaussian shape but which duration is now

$$T_{out} = T_p \times [1 + \frac{(\frac{d^2\Phi(\omega)}{d\omega^2})_s^2}{4b^2}]^{\frac{1}{2}} \tag{7}$$

Figure 2.2.1 shows the variation of the output pulse duration with the input pulse duration, for the case of propagation of a gaussian pulse through 10

[3] see for ex. S. De Silvestri, P. Laporta and O. Svelto, IEEE J. Quantum Electron. QE-20, 533 (1984).

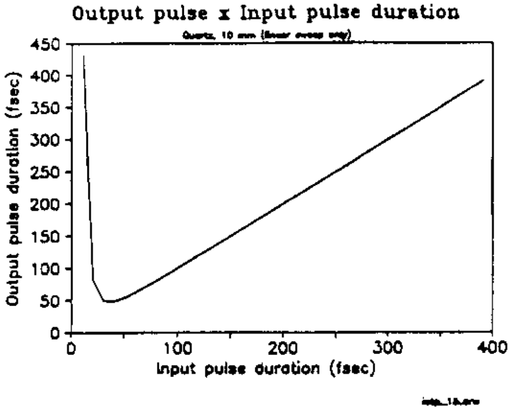

Figure 3: Duration of the output pulse as a function of the input pulse for propagation of a gaussian pulse through 10 mm of quartz

mm of quartz considering only the effects of the quadratic phase distortion. It can be seen that, in this example, the distortion becomes noticeable as the input pulse duration T_p becomes shorter than 50 femtoseconds becoming very severe for pulses sortet than about 25 femtoseconds. Notice that the numbers chosen for this example are typical in many laboratory situations.

2.2.2 Output pulse frequency sweep

The output pulse described by equation (4) contains also a time dependent phase term, given by equation 6, which describes a frequency sweep across the duration of the pulse. To obtain the instantaneous frequency we differentiate the phase (in the time domain) to obtain:

$$\omega(t) = \frac{d[\omega_0 t - \phi(t)]}{dt}$$

$$\omega(t) = \omega_0 + \frac{2\frac{d^2\Phi(\omega)}{d\omega^2}}{2\frac{d^2\Phi(\omega)}{d\omega^2}^2 + 8b^2} \times t. \quad (8)$$

(9)

Equation (9) indicates that the instantaneous frequency of the output pulse (equation (4)) increases linearly with time, what is called a positive frequency chirp. Note that here the frequency chirp is not associated with any increase in the bandwidth of the propagating pulse, since we assumed only dispersive effects. This is in opposition to the situation in which an intense pulse undergoes the process of Self Phase Modulation (SPM) caused by the modulation of the refractive index and actually generates new frequencies in its spectrum, increasing its own bandwidth.

The chirp acquired by the pulse due to dispersion can always be exactly compensated, by propagating the pulse through a medium (or system) which has the opposite type of phase distortion, thus regenerating exactly the initial pulse.

2.3 Chirped pulse propagation

For a gaussian pulse which has a linear frequency sweep

$$e(t) = exp[-(2\ln 2)(\frac{t}{T_p})^2] \times exp[j(\omega_0 t + \frac{\delta\omega}{2T_p}t^2]) \quad (10)$$

the output pulse can also be calculated in closed form, following the same steps described above. In this case a more general expression for the output pulse duration results which is

$$T_{out} = T_p \times [(1 - (\frac{d^2\Phi(\omega)}{d\omega^2})_s \frac{\delta\omega}{T_p})^2 + \frac{(\frac{d^2\Phi(\omega)}{d\omega^2})_s^2}{4b^2}]^{\frac{1}{2}}. \quad (11)$$

This equation shows that according to the relative signs of the group delay dispersion, $\frac{d^2\Phi(\omega)}{d\omega^2}$, and of the frequency sweep coefficient, $\delta\omega$, the pulse can be made longer or shorter after propagation. When the chirp is positive, $\delta\omega > 0$, as for the case of the pulse described in equation(4), propagation through a medium with positive group delay dispersion will increase the pulse duration. On the reverse, if the chirp is negative, the pulse duration will be shortened until it reaches a minimum duration and further propagation will lead to broadening. At the point of minimum duration the frequency sweep is nulled, and the pulse is transform limited. Figure 2.3 shows the measured pulse duration for a pulse propagating through a variable path of SF10 glass. A clear minimum is noticeable, and this technique is widely used in laboratories to identify any frequency sweep in the pulses.

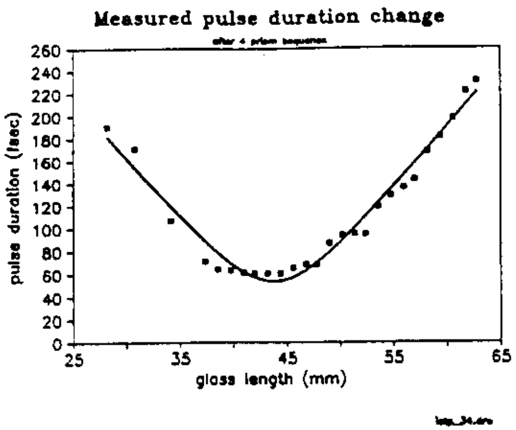

Figure 4: Measured and calculated pulse duration for chirped pulse propagation through a variable length of SF10 glass.

3 Linear sweep compensation.

In the visible region of the spectrum, the compensation of chirp acquired after propagation through material media requires the use of negative group delay dispersion. However, most material media produce positive group delay dispersion in this spectral range, so that it is necessary to resort to special devices which make use of **geometric dispersion**[4] to achieve overall negative group delay dispersion. The more important setups for this purpose are:

- a diffraction grating pair;
- a prism pair.

[4] See the notes by O.E. Martinez in this same Winter College on Ultrafast Phenomena.

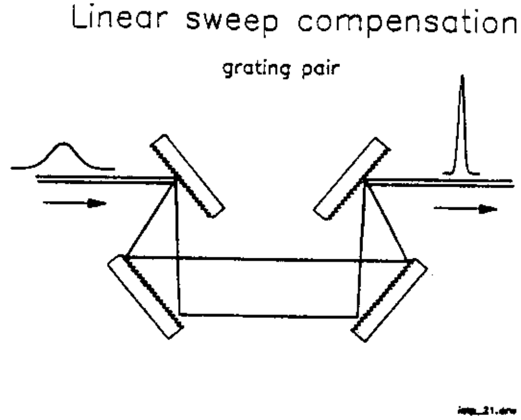

Figure 5: Grating pair in double pass configuration.

3.1 Grating pair

For a grating pair the group delay dispersion can be obtained from the original work of Treacy[5]. The expression is

$$\left(\frac{d^2\Phi(\omega)}{d\omega^2}\right)_g = \frac{-4\pi^2 c l_g}{\omega^3 d^2[1-(\frac{2\pi c}{\omega d}-\sin\gamma)^2]} \tag{12}$$

where c is the speed of light, d is the groove spacing of teh grating γ is the angle of incidence in the first grating and l_g is the grating spacing. For a grating with 600 lines per millimiter and incidence angle of 45°, at the wavelength of 620 nm the group delay dispersion is

$$\left(\frac{d^2\Phi(\omega)}{d\omega^2}\right)_g = -1710 \times l_g$$

for a single grating pair, with the grating separation expressed in centimeters and the group delay dispersion coefficient in fs^2. Most of the times the grating pair is used in a double pass configuration, as shown in Figure 3.1. Comparing with Figure 2 it can be seen that this ammount of group delay dispersion is equivalent to a negative quartz length of -3.8 cm. Figure 3.1

[5] E.B. Treacy, IEEE J. Quantum Electron. QE-5, 454 (1969).

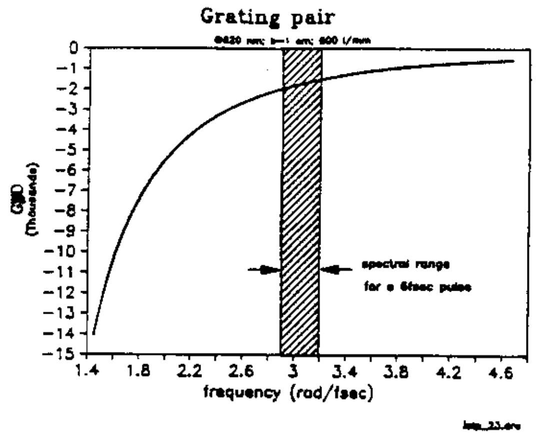

Figure 6: Group delay dipersion coefficient for a single grating pair.

shows the variation of the group delay dipersion coefficient for the gratings, $\frac{d^2\Phi(\omega)}{d\omega^2}_g$, as a function of the angular frequency. The shaded area indicates the FWHM of a 6 fs duration pulse. It can be seen that there is a noticeable cange of this coefficient within this range, pointing out to the fact that higher order dispersion terms become important at such short pulse durations. Grating pairs are currently widely used in pulse compressors[6] for pico and femtosecond pulse generation.

3.2 Prism pair

The prism pair, devised by R.L. Fork and O.E. Martinez at AT&T Bell Labs, became one of the more important dispersion compensation devices[7]. Their most clear advantage over the diffraction grating pair is the fact that their insertion loss can be very low, since Brewster angle prisms can be frequently used. However they cannot provide as much compensation as the grating pair. Their most important application is as intracavity dispersion compensators in 'colliding pulse mode locked lasers', which after

[6] See notes by A.M. Johnson in this same Winter College on Ultrafast Penomena.
[7] R.L. Fork, O.E. Martinez and J.P. Gordon, Opt. Lett. 9, 150 (1984).

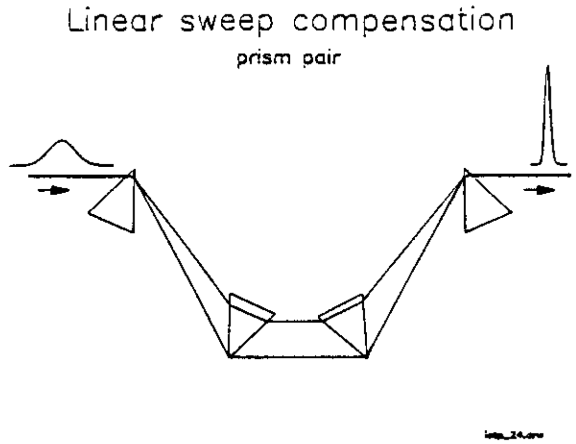

Figure 7: Four prisms sequence used as pulse compressor.

their introduction became able to generate pulses as short as[8] 27 fs. Figure 3.2 shows a typical four prism configuration. The ammount of group delay dispersion that can be contributed by the four prisms arrangement can be adjusted in two ways: either by changing the prism spacing, l_p, or by translating one of the prisms parallel to its base, inserting more or less glass in the path of the propagating pulse. Table 1 lists the values of the second derivative for some typical materials and systems. The value of $(\frac{d^2\Phi(\omega)}{d\omega^2})_p$

Table 1: Typical values for $\frac{d^2\Phi(\omega)}{d\omega^2}$ at 620 nm.

system	$\frac{d^2\Phi(\omega)}{d\omega^2}$ (fs^2)
quartz	$540 l_q$(cm)
BK-7 glass	690 l_b(cm)
double grating pair (600 lpmm)	$-3640 l_g$(cm)
prism pair (quartz, 60°)	650-$32 l_p$(cm)

[8] J.A. Valdmanis, R.L. Fork and C.V. Shank, Opt. Lett. 10, 131 (1985).

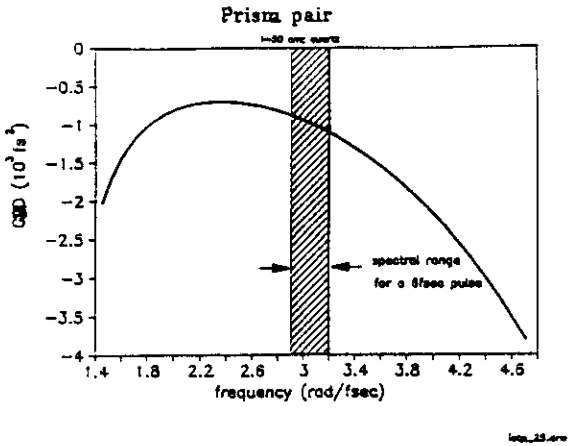

Figure 8: Calculated second order group delay dispersion for a double prism pair (quartz, 50 cm spacing, 60°).

for the prisms can be calculated from the propagation equations[9], and is shown in Figure 8 for the case of equilateral prisms made of quartz spaced by 50 cm.

It should be noted that here, the slope has the opposite sign as that of the grating pair (Figure 3.1), and in addition, there is a zero slope point in the infrared region of the spectrum, where the cubic term will be null.

4 Cubic phase distortion

The next important term in the Group Delay series expansion is the one related to $\frac{d^3\Phi(\omega)}{d\omega^3}$, which is responsible for generating a parabolic sweep of the group delay across the spectrum of the pulse (see Figure 1 and equation (2). For a positive value of $\frac{d^3\Phi(\omega)}{d\omega^3}$ this parabolic frequency sweep tends to cause a larger delay for the high and low frequency ends of the spectrum of the propagating pulse. Thus the associated distortion will no longer be

[9]R.L. Fork, O.E. Martinez and J.P. Gordon, Opt. Lett. 9, 153 (1984); O.E. Martinez, J.P. Gordon and R.L. Fork, JOSA A1, 1003 (1984); R.L. Fork, C.H. Brito Cruz, P.C. Becker and C.V. Shank, Opt. Lett. 12, 483 (1987).

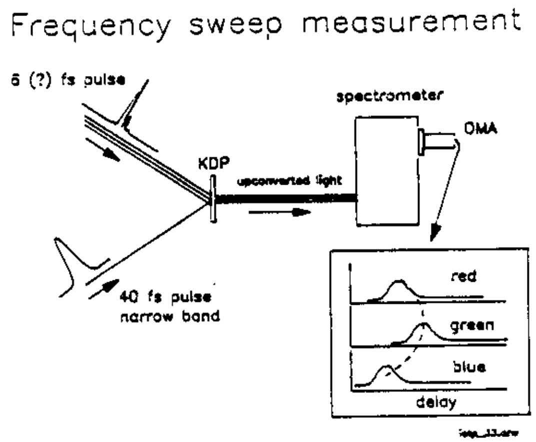

Figure 9: Experimental scheme for measuring the frequency sweep in a pulse.

symmetric, as for the second order term. The pulse will acquire a tail, and beating between the high and low frequencies will cause this tail to have fast oscillations. For the case of a negativeb $\frac{d^3\Phi(\omega)}{d\omega^3}$ the leading edge of the pulse will have the oscillating behavior. The importance of this term is relatively small, except in the case where the second order distortion is completely compensated. This is exactly the case in a pulse compression system, where usually a diffraction grating pair is used to compensate the linear sweep acquired upon propagation through an optical fiber. When the compressed pulse duration is below 10 fs, control of this parabolic sweep becomes esssential for generating a clean pulse, with sharp leading and trailing edges. For example, for a diffarction grating pair in a double pass configuration, the value of $\frac{d^3\Phi(\omega)}{d\omega^3}$ is $3120 l_g\ fs^3$ (@620 nm, 600 lpmm, l_g in cm), so that from equation (2) it can be found that the spread in group delay associated with this cubic term will be of the order of 15 fs.

This spread can be measured, using an upconversion technique in which a short pulse with 40 fs is mixed in a nonlinear crystal with the compressed pulse. The schematic of the measurement is shown in Figure 9. Figure 10 shows the time evolution of the frequency components of the compressed pulse for the case where the compressor uses a double pass through a grating

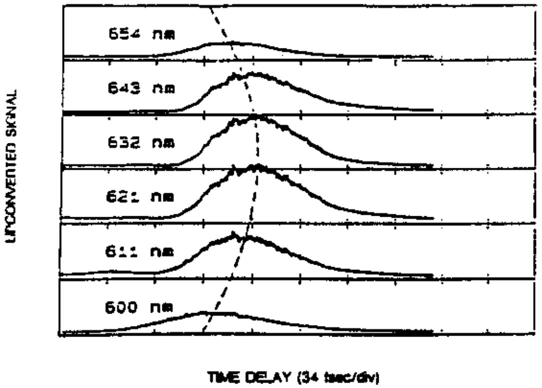

Figure 10: Time evolution of six frequency components of the pulse compressed with a grating pair, showing the parabolic frequency sweep.

pair to compensate for the linear sweep. It can be seen that a parabolic sweep occurs, with both the red and the blue side of the spectrum lagging behind its central portion.

From the phase shift introduced by the grating pair one can obtain the value of $(\frac{d^3\Phi(\omega)}{d\omega^3})_g$. Its value is always positive, as can be noted from the slope of the plot of $(\frac{d^2\Phi(\omega)}{d\omega^2})_g$ shown in Figure 3.1. For material media the value of $(\frac{d^3\Phi(\omega)}{d\omega^3})_m$ is also positive throughout te visible range of the spectrum (Figure 2). Thus the contribution from the grating pair and that from any material media in the path of the beam always add up, making the distortion worse. However the prism pair can be shown to have a third order term, $(\frac{d^3\Phi(\omega)}{d\omega^3})_p$, that can be either positive or negative, according to the particular geometry of the setup. For example, from Figure 8 it can be noted that the slope is opposite to that for the grating pair (Figure 3.1). This points to the conclusion that we can use both gratings and prism pair together, to obtain compensation both of the cubic and of the quadratic distortion. Table 2 lists the value of the third derivative for soma systems and materials of interest. Using the data in Table 1 and Table 2 a system can be designed such that it will provide the desired value of $\frac{d^2\Phi(\omega)}{d\omega^2}$, and

Table 2: Typical values for $\frac{d^3\Phi(\omega)}{d\omega^3}$ at 620 nm.

system	$\frac{d^3\Phi(\omega)}{d\omega^3}$ (fs^3)
quartz	240 l_q(cm)
BK-7 glass	332 l_b(cm)
double grating pair (600 lpmm)	3120 l_g(cm)
prism pair (quartz, 60°)	277-49 l_p(cm)

null the value of $\frac{d^3\Phi(\omega)}{d\omega^3}$, thus minimizing the distortion and, at the same time compensating any linear sweep the input pulses might have. Thus composite setup has been used to generate pulses with only 6 fs duration[10]. The time evolution of the frequency components from the compressed pulse obtained using this scheme is shown in Figure 11, which should be compared to Figure 10. It can be seen that here the several frequencies line up almost perfectly, to within the time resolution of this kind of measurement. The corresponding interferometric autocorrelation is shown in Figure 12.

5 Conclusion

We have discussed the effects of dispersion on ultrashort laser pulses, and shown that for the generation of clean sharp pulses dispersion control is mandatory. Grating pairs and prism pairs are the devices of choice for performing the phase correction. With the use of a compressor based on a prism pair and a grating pair pulses as short as 6 fshave been demonstrated. These pulses have opened an enormous range of applications in the study of dynamical processes in semiconductors and other condensed matter systems. Some of those applications are described in the following references:

1. C.H. Brito Cruz, R.L. Fork, W.H. Knox and C.V. Shank, 'Spectral hole burning in large molecules probed with 10 fs optical pulses',

[10] R.L. Fork, C.H. Brito Cruz, P.C. Becker and C.V. Shank, Opt. Lett. 12, 483 (1987); C.H. Brito Cruz, R.L. Fork, P.C. Becker and C.V. Shank, Opt. Lett. 13, 123 (1988).

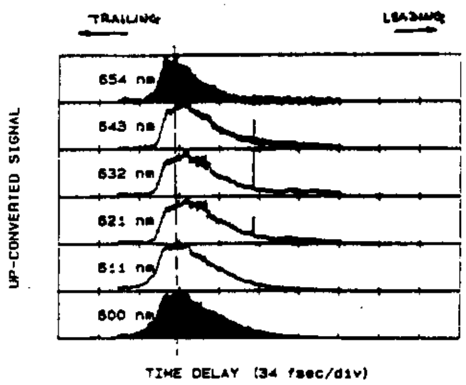

Figure 11: Time evolution of the frequency components from an optimally compressed pulse using a combination of gratings and prisms.

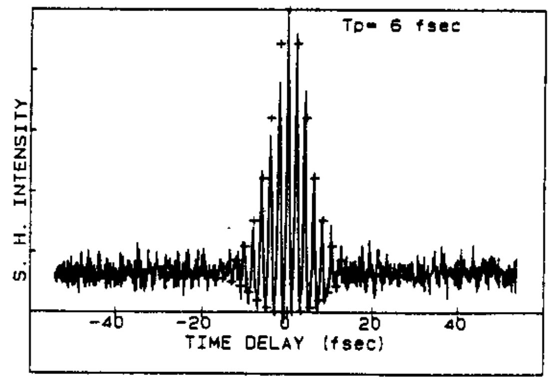

Figure 12: Measured interferometric autocorrelation for a 6fs pulse.

Chem. Phys. Lett. 132, 341 (1986);

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6 Useful references for further study

This is a list of papers covering the subject discussed here. (The list is not exhaustive.)

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