Measuring ultra short laser pulses

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Ultra short pulses (the order of a few fs) pulses allow time-resolved studies at the timescale of atom movement. These observations had never been available for a few years in the optical range. At FLASH these measurements are possible even in the soft X-Ray range, thanks to the effort of various scientists, working in different areas. One of these areas is the measurement of the pulselength of the optical laser used for some of the pump probe experiments carried out at FLASH.

During my stay here at DESY I worked most of the time at FLASH's laser hutch in the experimental area. I spent my time there setting up an autocorrelator that was supposed to be broken down, since there was no real signal coming out of it. After reading the manual, I could get a general understanding of what an autocorrelation function (ACF) is and a good understanding of the internal setup of this device. In addition I could get a very good ACF is from this machine. The next step was to design a remote control so one could use this machine without being inside the laser hutch. It has been a rather complicated thing because of the little troubles that arise before even being able to communicate with this device. The people that sold this device to DESY did not help us, and their manual has enormous omissions when explaining how to communicate with the autocorrelator via a PC. By a trial/error procedure I was able to communicate with this device and afterwards I started to write the real program that controlled this machine. I wrote it in LabView as it is standard in the group I was working in. This was a very educating time, as I was completely new to this programming environment. Finally we made some improved measurements using the program I made in LabView, but unfortunately it seems that either this autocorrelator is no reliable for such precise measurements (below tenths of femtoseconds), because its measurements vary in the order of 10-20 fs even with the same conditions.

Why an Autocorrelator?

In the measurement of ultra short laser pulses (lengths in the order of femtoseconds) the usual electronics become too slow to make any real time measurement. So the experimentalists have to come up with a way to measure such kind of laser pulses. One of the most popular ideas so far is the use of an autocorrelator, using short light pulses to measure short laser pulses. This is not just obstinacy, since the usual time response of the electronics, even in optical arrangements such as photodiodes, is of the order of 20 picoseconds

The SHG crystal is used in this field because it is a nonresonant process of electronic origin, so the nonlinear response from the crystal is fast enough to measure pulses of few times 10^{-15}s, let's remember that 10^{-15}s is the Bohr's period of a valence electron.

The design for the autocorrelator, regardless of the rather peculiar name,is quiet simple. It consists of a Michelson interferometer combined with a second harmonic generator (SHG) crystal in the place where the usual detector should go.

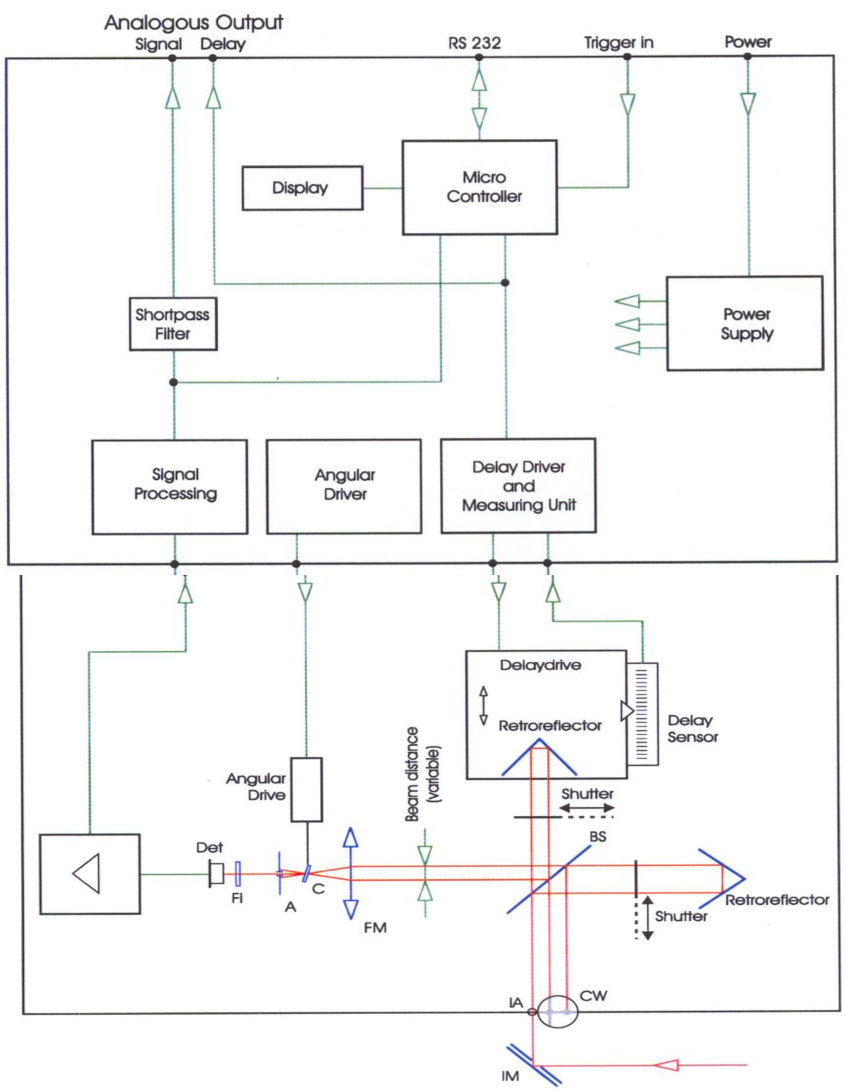

Figure 1

IM – Input Mirror
IA – Input Aperture
CW- Control Window
BS - Beam Splitter
SH - Shutter

FM – Focus Mirror
C - SHG-Crystal
A - Aperture
Fi - Filter
Det - Detector

The displacement of the scanning retro reflector is measured, and along with the settings for the scan range and the intensity of the light emitted by the SHG crystal, they are fed to the machine electronics to calculate the autocorrelation function.

The autocorrelation function, the measuring conditions and the calculated parameters (such as the full width at half maximum FHWM) can be retrieved by a PC. Nevertheless correct alignment can online be made at the optical unit's box. This is essential to get a good signal out of the autocorrelator, since a trial error procedure varying the angle of the crystal (it varies the optical axis's orientation), the beam distance and the focus distance is the only way of maximizing the signal strength. As a side product of this knowledge acquiring process I made a "Checklist" for the future users of this device, in order to make it easier and faster for them to use the autocorrelator.

What does an autocorrelation function measures?

The term autocorrelation is used when doing a cross correlation of a signal with itself. The cross-correlation function between two arbitrary functions *f* and *g* is defined as follows:

$$(f * g)(x) \equiv \int f^*(t) g(x+t) dt \quad , \tag{1}$$

where the integral is over the appropriate values of t, and f^* stands for the complex conjugate of *f*.

The cross-correlation is similar in nature to the convolution of two functions. Whereas convolution involves reversing a signal, then shifting it and multiplying by another signal, correlation only involves shifting it and multiplying (no reversing).

In an autocorrelation, since it is the cross-correlation of a signal with itself, there will always be a peak at a lag of zero.

For example, consider two real valued functions f and g that differ only by a shift along the x-axis. One can calculate the cross-correlation to figure out how much g must be shifted along the x-axis to make it identical to f. The formula essentially slides the g function along the x-axis, calculating the integral for each possible amount of sliding. When the functions match, the value of the cross-correlation is maximized. The reason for this is that when positive peaks are aligned, they contribute to making the integral larger. Even , when negative peaks (negative areas) align, they also make a positive contribution to the integral because the product of two negative numbers is positive.

With complex valued functions f and g, taking the conjugate of f ensures that aligned lumps (or aligned troughs) with imaginary components will contribute positively to the integral.

At the crystal the two beams interfere producing the second harmonic (this is why it is necessary the SHG crystal), but due to the different paths this procedure will vary its intensity. The real electric field on the detector, is the resulting interference between E_1 and E_2 this is

$$E(t) = E_1(t) + E_2(t+\tau) \qquad \textbf{(2)}$$

And the intensity at this point is given by the electric field squared averaged over one light period T

$$I(t,\tau) = \varepsilon_0 c n \tfrac{1}{T} \int_{t-T/2}^{t+T/2} \left| E_1(t'-\tau) + E_2(t') \right|^2 dt' \qquad \textbf{(3)}$$

The actual signal recorded at the output of the interferometer is the intensity, Ĩ, averaged over the response time τ_{res} of the scan. In the case of ultrashort pulses τ_{res} is several orders of magnitude bigger than τ_p (the pulse duration) and what is being measured is the time integral $\int_{-\infty}^{\infty} I(t',\tau)dt'$.

To a complex electric field E(t) corresponds an intensity autocorrelation function defined by:

$$A(\tau) = \int_{-\infty}^{\infty} I(t) I(t-\tau) dt \qquad \textbf{(4)}$$

The two parallel beams with a variable delay are generated, then focused into the second-harmonic-generation crystal. From (3) Only the beam propagating on the optical axis, proportional to the cross-product E(t)E(t − τ), is retained. This signal is then recorded by the detector, which measures

$$I_s(\tau) = \int_{-\infty}^{\infty} \left|E(t)E(t-\tau)\right|^2 dt = \int_{-\infty}^{\infty} I(t)I(t-\tau)dt \quad (5)$$

$I_A(\tau)$ is exactly the intensity autocorrelation given by (4) multiplied by a proportionality factor in which we are not interested for our measurements since the amplitude of a pulse doesn't change its FWHM.

The intensity autocorrelation width of a pulse is related to the intensity width. In our measurements a Gaussian time profile was assumed so , the autocorrelation width is $\sqrt{2}$ longer than the width of the intensity. This measurement can be made in the controls unit of the autocorrelator using cursors to measure b, B and H (see figure 2). Due to the automatic scan control the complete width B is defined by the actually selected scan range. Therefore the autocorrelation FWHM is calculated as:

$$FWHM = \frac{b \times Scanrange}{B} \quad (6)$$

To get the real pulse duration, as stated above, one has to correct the ACF width with a form factor depending on the pulse shape.

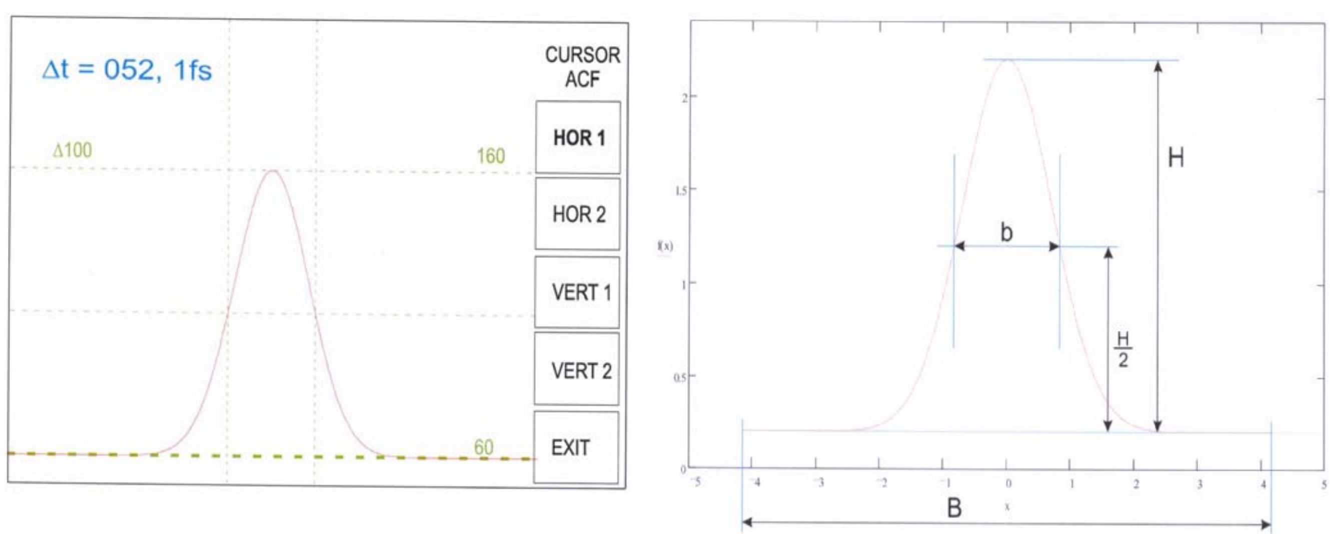

Figure 2

Since I was working on the remote controls for this machine and to get a more reproducible measurement, I managed to save the ACF to a file and then fit it assuming a Gaussian profile with Origin 7.5 . The laser we used for these trial measurements has the following expected characteristics (each pulse):

Pulse energy ~ 1 nJ

Pulse time length ~ 70 fs FWHM

Pulse spectralwidth ~ 20 nm FWHM

Central wavelength = 800 nm

Repetition rate= 108MHz

Due to the optical crystal (pockels cell) the spectralwidth as well as the central wavelength have a variation of 5 nm.

Results

With aid of the Origin, I could fit the ACF to a Gaussian and assuming that the pulse was a Gaussian of the form

$$f(x) = Ae^{-2\frac{(x-x_0)^2}{\sigma^2}} \qquad \textbf{(7)}$$

where *A* is the amplitude, x_0 is where the pulse is centered and σ is the standard deviation. The relationship between FWHM and σ is :

$$FWHM = \sqrt{2\ln(2)}\sigma \qquad \textbf{(8)}$$

Since this is only the ACF FWHM (the intensity autocorrelation) in arbitrary units, we still need to scale it then divide by $\sqrt{2}$ to get the pulse time- length .

In short, if we call τ the pulse length we have:

$$\tau = \frac{\sqrt{2\ln(2)}\sigma \times Scanrange}{Samplesize(256)\sqrt{2}} = \frac{\sqrt{\ln(2)}}{256} \times \sigma \times Scanrange \cong 0.0032521 \times \sigma \times Scanrange \qquad \textbf{(9)}$$

With this information I got the following graph for the intensity ACF:

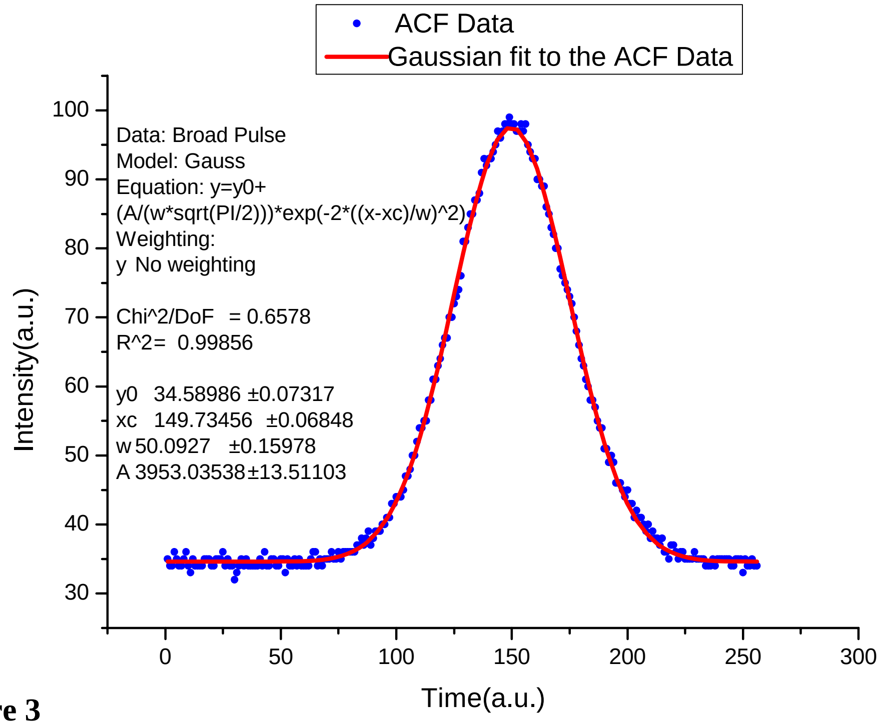

Figure 3

Using the fitting made by Origin and applying (9), I calculated the following pulse duration for this laser.

Pulse length ("broad" laser pulse) = 343.3 $\pm$ 1.0 fs

Afterwards we repeated the measurement without some of the optics that "elongated" the leaser pulse, this way we used a more "pure" laser pulse which was indeed much shorter than the first one (see figure 4).

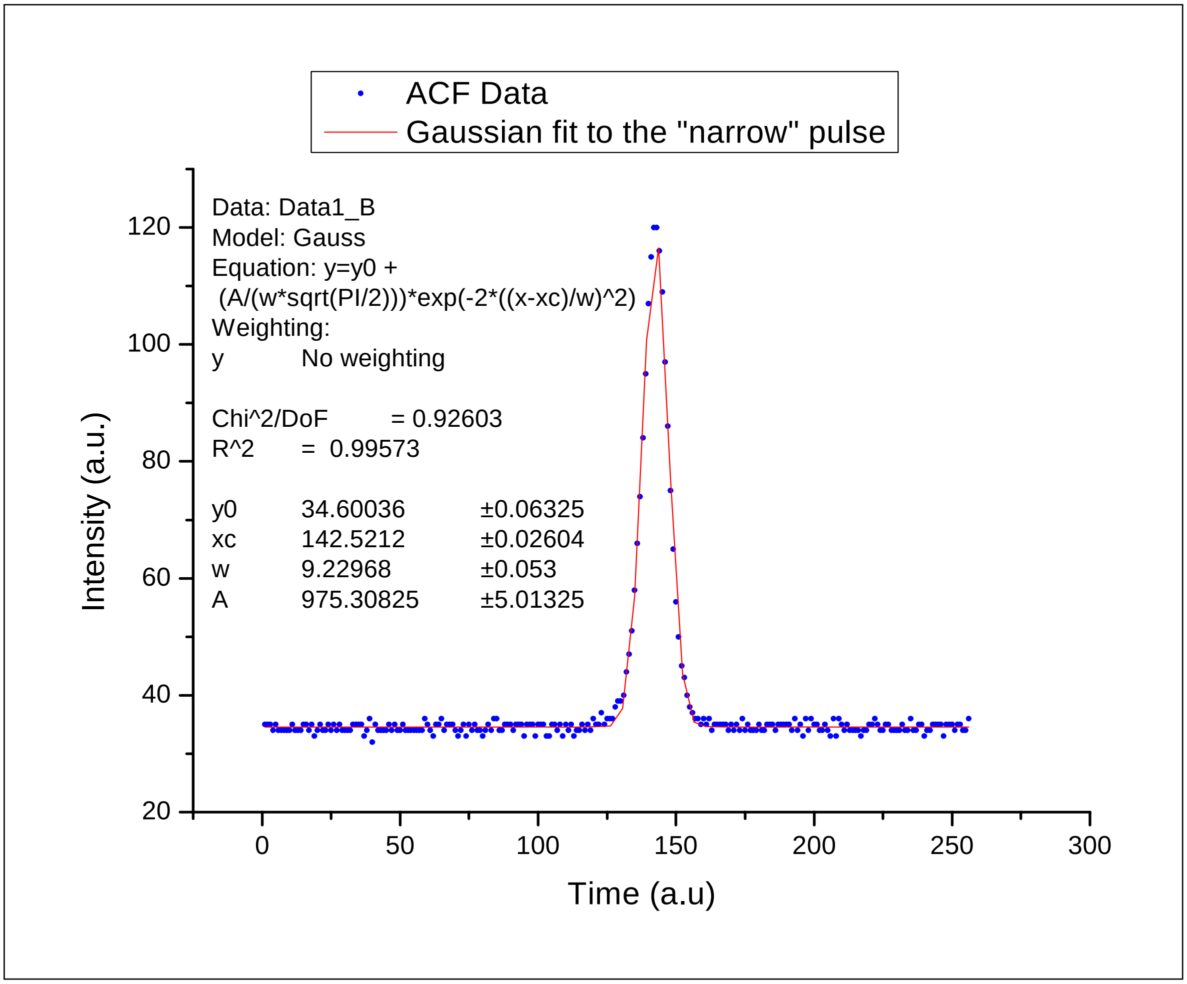

Figure 4

This time the laser pulse's time length was:

Pulse length ("narrow" laser pulse) = 63.2 ± 0.3 fs

As it can be seen from the graphs the assumption of the pulse's shape has been quite good. It would have been nice to get some other data or make a statistical study of the measured pulse length but the time at the laser hutch was limited, so these other improvements should be made in the future.

In this two results I am writing the fitting uncertainty, although the systematic error reported by the autocorrelator's manufacturer is of 1 fs in this Scanrange.
In the last measurements we saved many ACF functions to a file, which provided me with sufficient data to make a statistical analysis of the reported laser pulse duration. And I obtained the following results:

Pulse length (rms): 63.35 fs
Standard deviation: 0.49 fs
Averaged pulse length: 63.38 fs

Anyhow, the systematical error of 1 fs so we can be very confident that this results reflect the actual pulse length of the laser.

REFERENCES

Figures 1 and 2 were taken from the autocorrelator's manual (a "Pulse Check" autocorrelator sold by APE GmbH) as well as some information regarding the machines operation.

All theory comes from:

Diels, Jean Claude; Rudolph, Wolfgang, "Ultrashort Laser Phenomena", Academic Press, USA 1996.