Ultrafast Optical Pulses

Lisa M. Larrimore

Ultrafast pulses on the femtosecond time scale allow fundamental processes, such as atomic transitions, to be measured. To investigate the formation of optical pulses, an argon ion laser was used to pump a titanium-doped sapphire laser. The mode-locked output of this laser was measured with a spectrometer and an autocorrelator to find a frequency pulse width of $\sigma_\omega = 86 \pm 3$ rad/ps and a temporal pulse width of $\sigma_t = 14 \pm 1$ fs. The Heisenberg uncertainty principle demands that $\sigma_\omega \sigma_t \geq 1$, where the minimum uncertainty corresponds to a transform-limited pulse; for this investigation, $\sigma_\omega \sigma_t = 1.2 \pm 0.1$, so our pulse was nearly transform-limited. This conclusion is supported by overlaying the measured autocorrelation function with one calculated from the frequency spectrum, assuming a transform-limited Gaussian pulse; the two curves are nearly identical.

Introduction

Ultrafast optical pulses occur on a picosecond or shorter time scale, and pulses as short as six femtoseconds (6×10^{15} s) have been produced [1]. Because the energy in the amplifying material is released in such short pulses, each pulse has a very high power output. Peak powers of up to one terawatt (10^{12} W) have been achieved [1].

High energy pulses on the fs time scale allow many fundamental processes to be measured, such as the transfer of energetic excitations along the DNA molecule, the supersonic collapse of sonoluminescing bubbles, and the fundamental scattering processes in semiconductors [2]. Ultrafast pulses also have various technological applications, from microsurgery on the eye to ultrafast opto-electronics [1].

In order to generate ultrafast optical pulses, we used a titanium sapphire laser, since its large number of frequency modes allow it to produce a pulse on a very short time scale. We measured the frequency width of the pulse using a simple spectrometer, and we measured the temporal pulse width using an autocorrelator. The autocorrelator split the beam in two and inserted a separation τ between the two beams before recombining them in a crystal that displays simple harmonic generation (SHG), which means it can emit light at twice the frequency of the input.

By measuring the pulse width in both frequency and time, we can compare the results to the Heisenberg uncertainty principle, which limits the minimum uncertainty in these variables.

Theory

The energy-time uncertainty principle is given by

$$\Delta E \Delta t \geq \frac{\hbar}{2}, \tag{1}$$

where ΔE is the standard deviation in the energy and Δt represents the amount of time it takes the expectation value of some operator to change by one standard deviation [3]. Since for photons, $E = \hbar\omega$, Eq. (1) can be written as

$$\Delta\omega \Delta t \geq \frac{\hbar}{2}. \tag{2}$$

We thus see that a smaller Δt demands a larger $\Delta\omega$, or frequency range. This can be more rigorously shown using Fourier decomposition, resulting in the description "transform limited" for a pulse with the minimum temporal width allowed by Eq. (1) [2].

In order to generate ultrafast pulses, it is thus necessary to use a laser with a large gain bandwidth and a large number of longitudinal modes. This is why

the titanium-doped sapphire (Ti:Al_2O_3) laser is used: it has a gain bandwidth of about 300 nm and over 10^6 modes in a typical laser cavity [2]. An Argon laser, for comparison, only has about 12 modes [5], and a HeNe laser has only a few [2].

By bumping a component in the laser, such as the prism, we are able to initiate a spike. When that spike passes through the gain medium it takes out a lot of photons, depleting the gain until the spike comes back through the cavity. This gain-depletion is not sufficient to maintain mode-locking, but it is aided by the Kerr lens effect. This effect occurs when an intense beam of light increases the refractive index in the center of the beam, causing the beam to focus to a smaller point so that more of the pulse gets into the high-gain region. This allows the laser to become mode-locked, which means there is a fixed phase relationship between the different allowed modes.

Now consider the form of the wave packet output of a mode-locked laser. Since the "minimum uncertainty" wave packet in position-momentum space is a Gaussian, we consider this form when examining a temporal envelope [4]. We can write the field of our pulse as a function of time as

$$E(t) = E_0 \exp\left(-\frac{t^2}{2\sigma_t^2}\right) \exp\left(-i\omega_0 t\right). \tag{3}$$

The full width at half maximum (FWHM) of the intensity of this field is given by [2]

$$t_{FWHM} = 2\sqrt{\ln 2}\sigma_t. \tag{4}$$

The Fourier transform of a Gaussian pulse is also Gaussian,

$$E(\omega) = E_0 \exp\left(-\frac{\left[\omega - \omega^2\right]^2}{2\sigma_\omega^2}\right), \tag{5}$$

with a FWHM given by

$$\omega_{FWHM} = 2\sqrt{\ln 2}\sigma_\omega. \tag{6}$$

If we let $\Delta\omega = \sqrt{\langle\omega^2\rangle - \langle\omega\rangle^2}$, where $\langle\omega^2\rangle = \omega_0^2 + \sigma_\omega^2/2$ and $\langle\omega\rangle^2 = \omega_0$, we find that $\Delta\omega = \sigma_\omega/\sqrt{2}$. The same equations hold for Δt, and so Eq. (2) becomes [2]

$$\Delta\omega\Delta t = \frac{\sigma_\omega\sigma_t}{2} \geq \frac{1}{2}, \tag{7}$$

which means that

$$\sigma_\omega\sigma_t \geq 1. \tag{8}$$

Eq. (8) becomes an equality for a transform-limited pulse.

To compare our results with those predicted by the uncertainty principle, we wanted to measure the ultrafast pulse width in both frequency and time. The frequency width could be measured using a spectrometer, but electronics are too slow to measure something on the femtosecond time scale. To find Δt it was thus necessary to use an optical method. Simply measuring the interference can be problematic, since it is difficult to tell whether an interference pattern is from a dispersion-free pulse or not. Instead of using an interferometer to measure the first-order field correlation function, we used an autocorrelator to measure the second-order intensity correlation function. Even if pulses are dispersed (and thus not Fourier transform limited), measuring the intensity autocorrelation function still gives the proper signal [5].

In order to measure this function, we use an optical effect known as second harmonic generation (SHG), which utilizes the fact that there is some probability that an atom will absorb two photons of frequency ω and emit one photon of frequency 2ω. This can only be done with a non-centrosymmetric material, since if changing the direction of the electric field only changes the direction of the polarization, then the even powers of E in the expansion of P must go to zero, and it is the first of these even powers that results in SHG. The autocorrelator splits the field into two and delays one of the pulses by a time interval τ before recombining them and sending

them through the crystal. We then consider the second term in the expansion of the polarization inside the crystal,

$$P(t) = \chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \cdots , \tag{9}$$

where $\chi^{(i)}$ are the nonlinear optical susceptibility tensors [2]. By integrating the time averaged intensity, which is proportional to the square of the dipole moment, it can be shown that the autocorrelation signal is given by

$$\left\langle I^{2\omega_0}(\tau)\right\rangle \propto \left[1 + \exp\left(-\frac{\tau^2}{2\sigma_t^2}\right)\left[2 + \cos\left(2\omega_0\tau\right)\right] + 4\exp\left(-\frac{3\tau^2}{8\sigma_t^2}\right)\cos(\omega_0\tau)\right]. \tag{10}$$

From Eq. (10) it can be shown [2] that

$$\sigma_t = \frac{t_{FWHM-AC}}{2\sqrt{2}}, \tag{11}$$

where $t_{FWHM-AC}$ is the FWHM of the autocorrelation trace.

Procedure

A 4.5 W argon ion laser was used to pump a Ti:Al_2O_3 laser, which contained prisms to correct for the angular dispersion of the beam. When the Ti:Al_2O_3 laser was producing a stable mode-locked output, its spectrum and autocorrelation traces were measured. The spectrum was could be directly measured by sending the beam into a spectrometer whose output was sent to a digital scope and was compared to a spectrum analyzer. To measure the autocorrelation function, the beam was first sent through another pair of prisms to correct for dispersion and then to the autocorrelator. The beam was split using a 50/50 beam splitter, retroreflected off aluminum mirror corner cubes, and recombined with a different beam splitter. The two beam splitters ensured that each beam undergoes the same dispersion before recombining. A focusing mirror then directed the beam through a SHG crystal, which was then focused again through filters to remove the IR light and into a

photomultiplier tube (PMT). The output current of the PMT was converted to a voltage and read on a digital oscilloscope.

Results and Discussion

Figure (1) shows the output of the spectrometer used to measure the frequency width of the wave packet.

$$\omega_{FWHM} = \frac{2\pi c}{\lambda_1} - \frac{2\pi c}{\lambda_2}, \tag{12}$$

where $\lambda_1 = 790.2 \pm 1.0$ nm and $\lambda_2 = 840.8 \pm 1.0$ nm are the wavelengths at half the maximum height. This gives $\omega_{FWHM} = 143 \pm 6$ rad/ps. Equation (6) can then be used to determine that $\sigma_\omega = 86 \pm 3$ rad/ps.

The second-order intensity correlation function, $g^{(2)}$, is shown in Figure (2). Knowing that each pair of fringes is separated by one period of oscillation, or $\lambda_0/c = 2.72 \pm 0.02$ fs, and counting the number of data points between fringes, it was determined that each data point corresponded to 0.041 ± 0.002 fs. This allowed the horizontal axis to be rescaled in fs for this graph. Since the FWHM was measured to be 950 ± 20 points, our calibration gives $t_{FWHM-AC} = 39 \pm 2$ fs. Using Equation (11), $\sigma_t = 14 \pm 1$ fs.

Multiplying σ_ω and σ_t to compare with Equation (8) gives $\sigma_\omega \sigma_t = 1.2 \pm 0.1$.

Using Eq. (10), the autocorrelation function can be calculated from the spectrum plot in Figure (1), assuming a transform-limited Gaussian pulse. Using our data, this curve was generated by a program written by C. H. Grossman and T. D. Donnelly [2] and is shown as the solid line in Figure (3). The circles in Figure (3) are the measured autocorrelation function from Figure (2). The two curves are nearly identical except at the edges, which means that our pulse was nearly transform-limited.

Conclusion

To explore the fundamentals of ultrafast pulse formation, pulses on the femtosecond time scale were produced by mode-locking a Ti:Al_2O_3 laser, which was pumped by an Ar ion laser. Using a spectrometer and a spectrum analyzer, the frequency width of the signal was measured to be $\sigma_\omega = 86 \pm 3$ rad/ps. By aligning an autocorrelator, the temporal pulse width was determined to be $\sigma_t = 14 \pm 1$ fs. The product of the laser pulse width in the frequency and time domains is constrained by the Heisenberg uncertainty principle, which can be expressed as $\sigma_\omega \sigma_t \geq 1$; multiplying our results gives $\sigma_\omega \sigma_t = 1.2 \pm 0.1$. Assuming a transform-limited Gaussian pulse, a theoretical second-order intensity autocorrelation function was calculated from the frequency spectrum and was seen to overlap nearly exactly with the measured autocorrelation function. From this graph and from our comparison of the pulse width with the Heisenberg uncertainty principle, we conclude that our pulse was nearly transform-limited.

References

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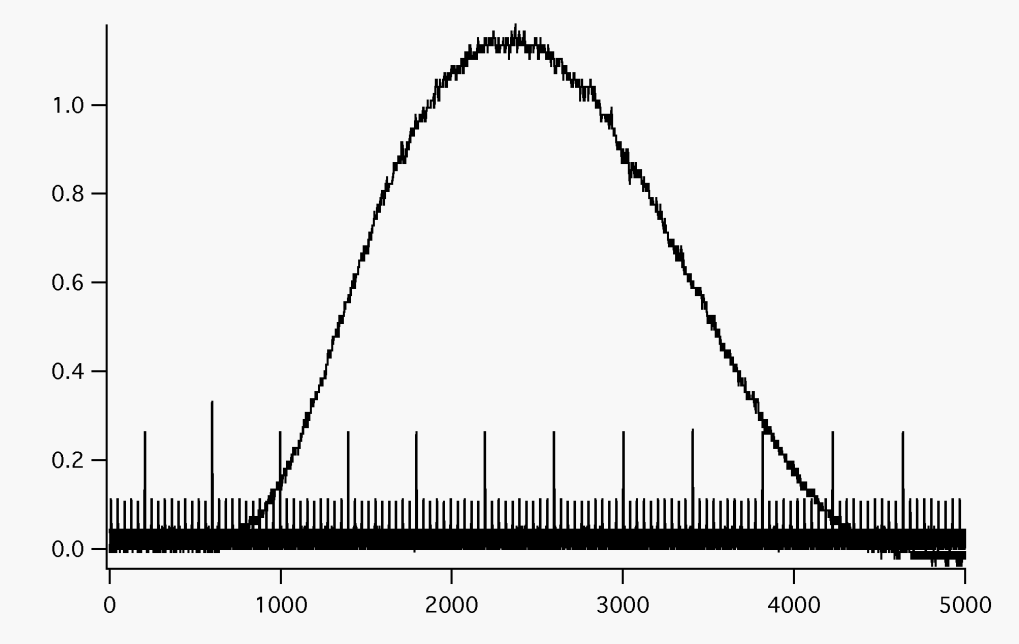

Figure 1: The curve is the spectrometer output, and the spikes from the spectrum analyzer provide a scale; the largest spike corresponds to 770 nm, and the other big spikes mark off 10 nm intervals. Using this scale, the wavelengths at half the maximum were determined to be 790.2 ± 1.0 nm and 840.8 ± 1.0 nm, resulting in $\sigma_\omega = 86 \pm 3$ rad/ps. The center wavelength is $\lambda_0 = 815.5 \pm 3.0$ nm. In this graph, the horizontal axis is labeled by data point number and can be ignored.

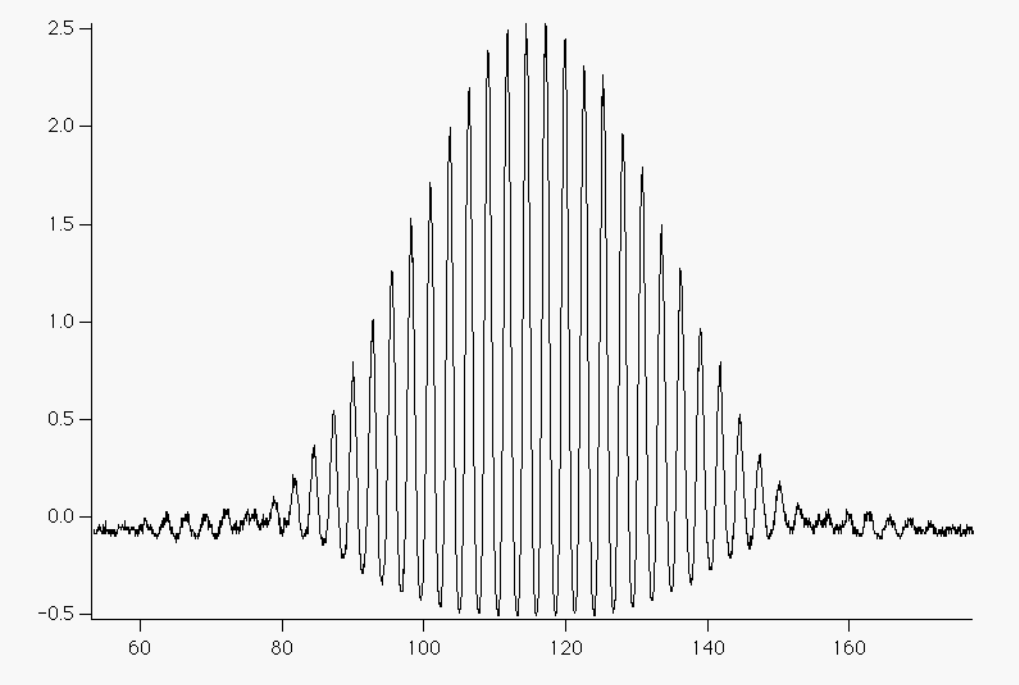

Figure 2: Second-order intensity autocorrelation signal from the autocorrelator. The horizontal scale in fs was calculated by finding the number of data points between fringes and knowing that each fringe separation is λ_0/c. The FWHM of this autocorrelation signal is $t_{FWHM-AC} = 39 \pm 2$ fs, resulting in $\sigma_t = 14 \pm 1 fs$.

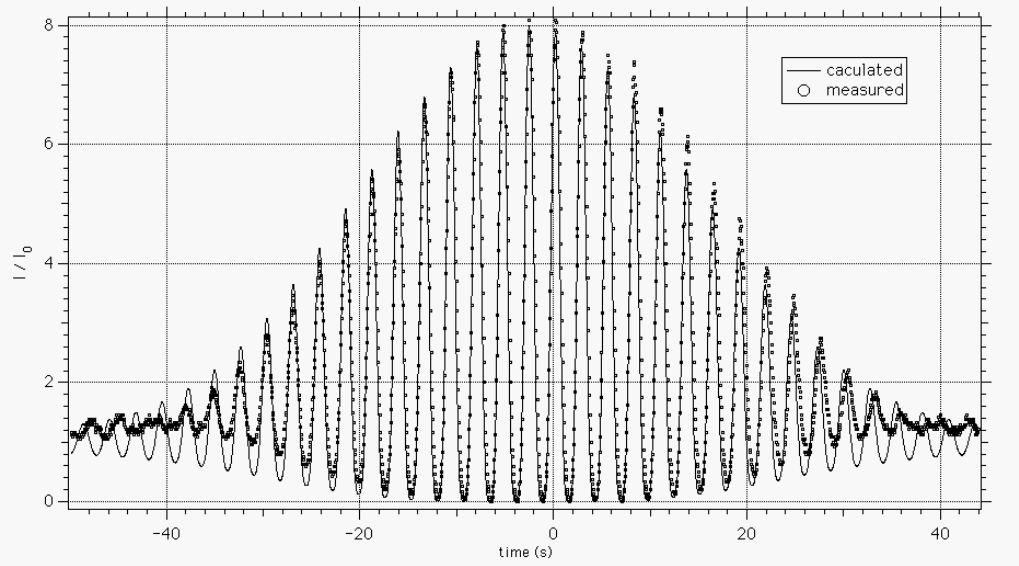

Figure 3: The solid line is the autocorrelation function calculated from the spectrum plot, assuming a transform-limited Gaussian pulse, and the circles are the measured autocorrelation function from Figure (2).