

Ultrafast Mirrors for Femto-second Pulse Duration laser Systems

Ryan McGuigan, MPhys

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1. Introduction

Advances in laser technology have paved the way for the development of powerful, ultrafast lasers with pulse lengths as short as 10fs or less^[1]. Building such systems has of course come with many challenges for the precision optics community. Laser systems functioning in the femto-second pulse duration regime suffer acutely from the effects of optical dispersion compared to those in the nano or pico-second regimes^[2]. As ultrashort pulses propagate through media they acquire large Group Delay Dispersion (GDD). This in turn causes pulse broadening, lowering the peak power of the pulse and limiting the duration of the output pulse as well as introducing a chirp that causes the instantaneous frequency to vary along the length of the pulse^[3]. Ultra-fast systems therefore require low GDD components to access and maintain femto-second pulse durations.

The ultra-short pulse regime is additionally characterised by completely different laser damage mechanisms. For pulses with a duration of 0.5ns or more, the LIDT mechanisms and the scaling of LIDT from one given pulse duration, beam diameter and wavelength to another is well understood. However for ultra-short pulses different mechanisms dominate, making it more difficult to fully understand the LIDT and related scaling factors in this pulse duration regime. This is, for example, illustrated by the experimentally observed deviation^{[4][5]} (in the ultra-short regime) from the well-known LIDT scaling law that states critical fluence scales with the square root of pulse duration^[6], $\tau^{1/2}$. The damage mechanism is thought to be related less so to the actual quality of the coating deposition process and more-so to the intrinsic properties of the coating materials such as the electron band-gap and therefore potentially sets upper limits on achievable fluences.

Additionally, the optics industry has had to keep up with and facilitate these developments by offering the means for tunable Group Delay Dispersion control. Ultra-fast optics are essential for pulse formation in femto-second solid state lasers in particular, where they are utilised in compressor systems to reduce pulse duration via a combination of positive chirping of a pulse and negative GDD components^[3]. This white-paper provides a brief overview of LIDT in the ultra-short pulse regime, the detrimental impact of dispersion in this regime and explores the design and capabilities of ultra-fast mirrors.

2. Why we need Ultra-fast Optics

2.1. Group Delay Dispersion

To understand why low GDD coatings are essential for laser systems consider the frequency spectrum of a pulse and the phase acquired when it passes a distance x through a medium of refractive index n . A typical pulse structure is given by the electric field (1)

$$E = E_0 e^{(-\Gamma t^2 + i\omega_0 t)} \tag{1}$$

Where Γ is the pulse shape factor and is inversely proportional to the square of the pulse duration ($\Gamma \propto \tau^{-2}$), and ω_0 is the central pulse frequency. The structure is essentially a Gaussian envelope over an oscillatory factor, represented by the real and imaginary parts of the exponent respectively. The Fourier transform with respect to time reveals the spectral structure of the pulse which in fact contains a spectrum of frequencies about the central frequency ω_0 . The full width at half maximum (FWHM) $\Delta\omega$ of this spectrum and the FWHM duration Δt are related via a relation analogous to Heisenberg’s Uncertainty Principle in Quantum Physics, that is:

$$\Delta t \Delta \omega \geq 0.441 \tag{2}$$

It is important to note that this bound depends on the exact pulse structure, with (2) corresponding to the spectral content of a Gaussian pulse. Table (1) contains minimum values satisfying this relation for given wavelengths, consequently illustrating the breadth of the broadband spectral content within femto-second pulses.

Wavelength and minimum spectral width							
Pulse Width	248nm	355nm	532nm	633nm	800nm	1064nm	1550nm
1ns	0.0001nm	0.0002nm	0.0005nm	0.0007nm	0.001nm	0.002nm	0.004nm
1ps	0.1nm	0.2nm	0.5nm	0.7nm	1nm	2nm	4nm
50fs	2nm	4nm	10nm	12nm	21nm	38nm	80nm
10fs	10nm	21nm	47nm	67nm	110nm	190nm	400nm

Table 1: Wavelength bandwidths for a variety of pulse widths and central wavelengths. Longer width pulses exhibit a spectrum very close to the central wavelength whereas femto-second scale pulses exhibit a very broad spectrum.

According to table (1), in the femto-second regime the pulse spectrum in frequency space becomes very broad, meaning that the pulse consists of many different wavelengths about the central wavelength and the structure is therefore very vulnerable to dispersive effects in media as opposed to the pico-second regime where the effects are very weak. This is essentially the same as the characteristic splitting of a beam of white light passing through a prism into visible frequency components. The most characteristic effects are pulse broadening and a linear “chirping” of the pulse. A pulse traversing a distance x in a medium will acquire a reduced Γ given by (3).

$$\Gamma \rightarrow \frac{\Gamma}{1 + 4(\Gamma k''x)^2} \quad k'' = \frac{\lambda^3}{2\pi c^2} \frac{d^2n(\lambda)}{d\lambda^2} \quad (3)$$

This equation implies an increased pulse duration or pulse broadening. One negative consequence of this is a reduction in the peak power of the laser pulse (i.e. The pulse energy is more spread out over time due to energy conservation, reducing the peak). The k'' is a material thickness independent quantity known as the "Group Velocity Dispersion" or GVD whilst $k''x$ is the Group Delay Dispersion (GDD) and is conventionally measured in units of fs². The imaginary phase factor for Gaussian pulse (1) is also perturbed and acquires a quadratic term in time leading to an instantaneous angular frequency that varies linearly with time (as opposed to being a constant ω_0). This is what is referred to as a "chirped" pulse where one wing of the pulse may look more red or blue and the other may look more blue or red.

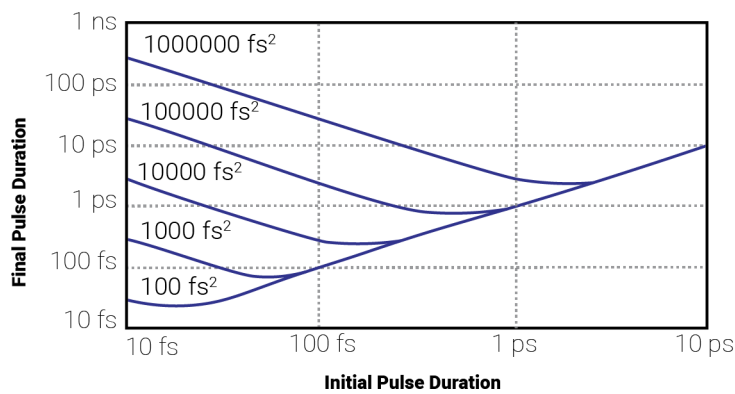


Figure 1: Pulse broadening as a function of input pulse duration. Note that even for very large GDD long pulses are barely affected by dispersion

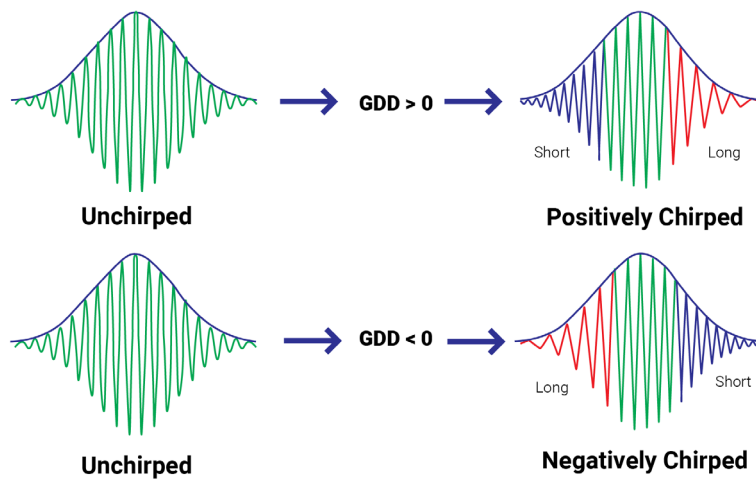


Figure 2: Illustration of pulse chirping. An unchirped pulse maintains a single instantaneous frequency (corresponding to the pulse wavelength) across its duration whereas a chirped pulse's instantaneous frequency slides from one wavelength to another along the duration.

A means to measure GDD is understandably vital in the manufacturing of ultra fast components. This is typically achieved through white light interferometry methods. This method is simple, versatile and does not take up much space. Incoherent white light is collimated towards a beam-splitter which directs beams towards a low-dispersion mirror and the optic being tested. The reflected beams then recombine and produce an interference pattern that is recorded by a computer. The dispersion of the test mirror is then extracted from this pattern^[7].

2.2. Laser damage in the ultra-short pulse regime

Over the last couple of decades much research has taken place to better understand the mechanisms that govern laser-induced damage threshold (LIDT) in the ultra-short pulse regime. As laser systems move towards more powerful sources and femto-second scale pulse durations any insight into the nature of LIDT in this less well-charted territory is critical.

Damage in the ultra-short regime for dielectric materials is thought to arise mainly due to the excitation of valence electrons to the conduction band typically via multi-photon absorption for more energetic photons (i.e. UV) or through Tunnel Ionisation for less energetic photons^[8] (i.e. IR) as opposed to mere heating and absorption due to intrinsic defects. These new conduction electrons may then absorb more photons and collide with valence electrons, ionising them if the conduction band electrons have absorbed enough energy from laser (known as electron impact ionisation). The valence electrons are hence excited to the conduction band, allowing them to absorb energy via free electron absorption and so on. The result is an exponential growth of conduction band electron density that, above a critical density 10^{21}cm^{-3} , causes the material to behave like a metal and acquire a non-negligible extinction coefficient. The huge amount of energy absorbed from the laser results in ablation of the coating as well as melting due to heating.

Much work was done by Mero et al. ^{[4][5]} with 1-on-1 tests to establish an (empirical) formula to predict the LIDT of commonly used dielectric coating materials in the fs-regime for single pulses. The suggested scaling is as follows:

$$F_{th} \approx (c_1 + c_2 E_g) \tau^k \quad (4)$$

Where F_{th} is the critical fluence, k is an empirical exponent ($k \approx 0.30 \pm 0.03$) E_g is the material bandgap, c_1 and c_2 are empirical factors ($c_1 \approx -0.16 \pm 0.02 \text{Jcm}^{-2}\text{fs}^k$, $c_2 \approx 0.074 \pm 0.004 \text{Jcm}^{-2}\text{fs}^k \text{eV}$). The factors c_1 and c_2 are empirical and only show a very weak dependence on film deposition and post deposition treatment. Having been determined from work based on oxide materials it is likely that they will change with other types of coating materials. The parameter k is thought to be pulse independent and only weakly material dependent. For commonly used coating materials, Mero et.al. give the material specific constants and electron band gaps. When inserted into the above formula, they suggest values for the critical fluence as per the table 2 below:

Material	n_{800}	E_g (eV)	k	F_{th} (Jcm ⁻²)
TiO ₂	2.39	3.3eV	0.28 ± 0.02	0.18
Ta ₂ O ₅	2.17	3.8eV	0.33 ± 0.02	0.30
HfO ₂	2.09	5.1eV	0.30 ± 0.02	0.49
Al ₂ O ₃	1.65	6.5eV	0.27 ± 0.02	0.67
SiO ₂	1.5	8.3eV	0.28 ± 0.02	1.11

Table 2: Theoretical critical fluences F_{th} for 800nm, 15fs pulses and measured k values of various thin film materials.

There is also evidence to suggesting the following empirical law^[9]

$$F_{th} = \frac{12Jcm^{-2}}{n^3} \quad (5)$$

Where n is the material refractive index. These empirical formulae have interesting consequences in the design of high LIDT optical coatings. They imply that a material dependent approach must be applied as the LIDT depends on fixed material properties like electron bandgap and refractive index, with (4) and (5) implying the larger the bandgap/lower the index the higher the LIDT. This means that a multilayer coating such as a broadband reflector may have a higher LIDT when produced with lower index materials, though this will have the effect of reducing the bandwidth of the high reflection zone. Perhaps most importantly this research suggests a natural upper limit to laser damage threshold as far as optical coatings are concerned; no matter how perfect the deposition process is the LIDT can only be so high!

3. Ultrafast Mirrors

The components of a laser system from mirrors to lenses to polarisers will all possess dispersive properties hence the more complex a femto-second laser pulse system is, the more time a pulse spends propagating through dispersive media and the worse it is distorted as a result^[9]. A typical ultra-fast system can easily contain a dozen or more mirrors. If these mirrors are dispersive then a pulse may become broadened and chirped to an unreasonable degree. Ultra-fast optics mirrors are a specialist line of products that are designed such that they possess dispersive properties that can correct for these effects. For example a pulse that has acquired strong positive GDD across its spectrum can have its GDD reduced by reflecting it off a highly negative GDD mirror. Mirrors can also possess a very low, constant GDD across their reflective bandwidths such that the dispersion experienced by a pulse can be minimised in comparison to a mirror that has not had its dispersive properties tailored in such a manner. It is also important that Ultrafast mirrors have as high a reflectivity as possible to reduce losses.

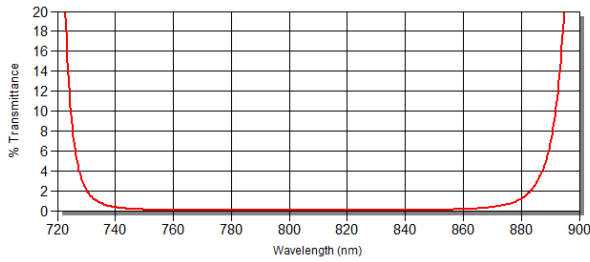
One of Manx Precision Optics' specialisations is custom low GDD, high LIDT, broadband mirrors. We have four lines of ultrafast mirror referred to as TTS, TTB, TTW and TTMH (standing for "Tunable Ti:sapphire", "Standard", "Broad", "Wide" and "Metal Hybrid").

3.1 Dielectric Mirrors

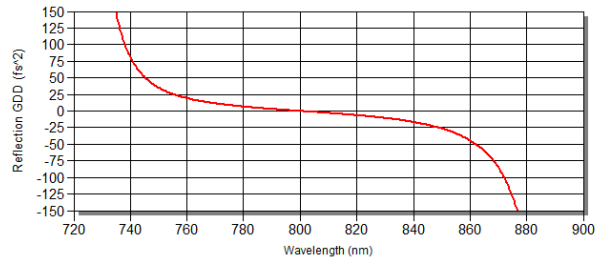
Dielectric Mirrors are very versatile for GDD control as they can be tailored across a range of wavelengths typically from the near and middle UV into the near Infrared (material dependent). Dielectric mirrors consist of many alternating dielectric layers (negligible absorption) of two or more refractive indices sitting atop a substrate and reflect light by utilising interference effects^[10]. Incident light deeply penetrates such a system which allows for tight control of the GDD with layer thickness directly corresponding to the phase and GDD introduced. Dielectric mirrors are often implemented in modern Ultra-fast laser systems over dispersive prisms or diffraction gratings as they are far less lossy, do not suffer from large cubic (and higher) order dispersions^[11] and are genuinely easier to install allowing for more compact systems.

3.1.1 Bragg Reflectors

The simplest dielectric mirror is the quarter wave stack consisting of a series of low and high index layers with individual thickness' equal to one quarter of the design wavelength. The result is virtually 100% reflectivity at the design wavelength as well as a highly reflecting band about this wavelength. The more layers the higher the reflectivity. Across this bandwidth the Group Delay varies slowly and linearly with wavelength making the Quarter Wave Stack ideal for maintaining or compensating for small, constant GDD.



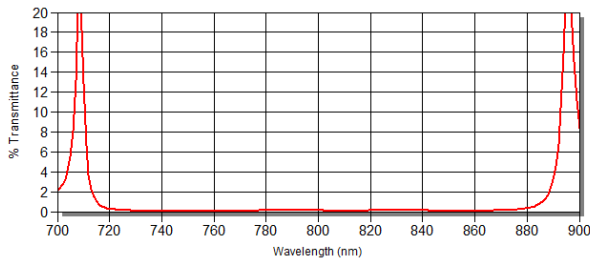
(a) Reflectivity of a standard (TTS) quarter wave Bragg reflector designed for an 800nm pulse wavelength. The LIDT is around $\geq 0.75 \text{Jcm}^{-2}$.



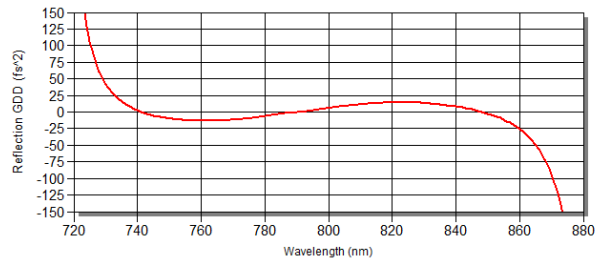
(b) Group Delay Dispersion of a standard quarter wave Bragg reflector designed for an 800nm pulse wavelength. The GDD is close to 0 at the design wavelength and varies slowly yet linearly from around 760nm to 840nm.

3.1.2 Broadband Mirrors (TTB and TTW)

The combination of sophisticated design methods such as Double Chirping and computer refinement techniques has allowed for the development of Ultrafast dielectric components with capabilities far beyond the standard reflector. Such specialist mirrors are essential for more demanding applications, for example a 5fs pulse corresponds to a 100THz, or 200nm bandwidth at 800nm and therefore requires a broadband mirror with enhanced operational wavelength, a simple Bragg reflector will not be suitable for this application.



(a) Reflectivity of a broadband (TTB) reflector designed for an 800nm pulse wavelength. Compared to the 800nm TTS this TTB Mirror boasts a bandwidth which is broader by about 40nm whilst maintaining a high LIDT ($> 0.45 \text{Jcm}^{-2}$ in 25fs). The TTW range is designed to cover even wider bandwidths (e.g 720-900nm)

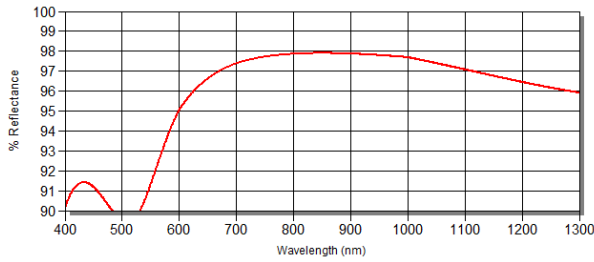


(b) Group Delay Dispersion of a broadband (TTB) reflector designed for a 800nm pulse wavelength. The GDD is very low across a broader bandwidth, only varying by $\pm 25 \text{fs}^2$ from 740nm to 860nm

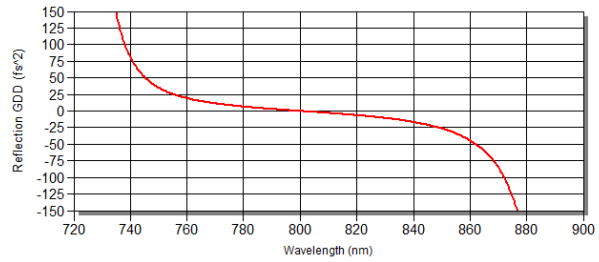
3.2 Metal Mirrors

3.2.1 Protected Metal

A single metal layer with a simple protective overcoat can be highly effective for Ultrafast purposes. A pulse does not have to penetrate far into a metal coating to be reflected hence GDD is low. Metal mirrors are simple and highly effective however the range of wavelengths and bandwidths over which they operate is limited in comparison to dielectric mirrors, and they are more vulnerable to degradation over time. In the nano-second regime they have poor LIDT due to high extinction coefficients, for example a silver metal mirror may have a typical LIDT of around 2.5Jcm^{-2} at 1064nm in 10ns whereas a dielectric mirror can withstand around $> 35 \text{Jcm}^{-2}$. On the other hand metal mirrors perform drastically better in the femto-second regime owing to the different damage mechanisms at play. According to a paper by Angelov^[12] a single layer metal mirror can have an LIDT that is approximately half that of dielectric mirrors under certain conditions.



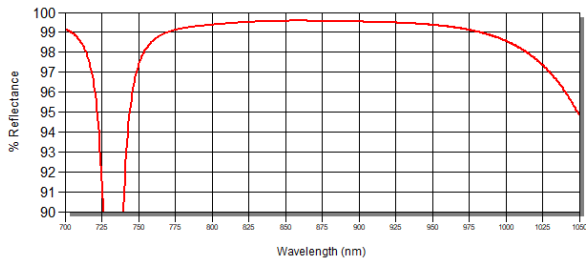
(a) Reflectivity of a protected silver mirror



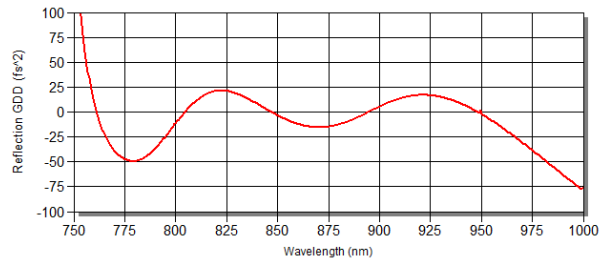
(b) Group Delay Dispersion of a protected silver mirror

3.2.2 Metal Hybrids (TTMH)

Metal hybrid mirrors combine the extremely low GDD and high reflectivity of metal mirrors with the durability and versatility of dielectric mirrors. These mirrors consist of a single metal layer overcoated with a dielectric stack, reinforcing the natural reflectivity of the metal with the interference properties of the dielectric layers. The result is an extremely broad reflectivity bandwidth with a much higher performance specification than an ordinary bare or protected metal mirror.



(a) Reflectivity of a silver hybrid (TTMH) mirror at 0 degrees



(b) Group Delay Dispersion of a silver hybrid (TTMH) mirror at 0 degrees

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