Design of dielectric high reflectors for dispersion control in femtosecond lasers

Róbert Szőcs, Ambrus Kőházi-Kis
Research Institute for Solid State Physics
H-1525 Budapest, P.O. Box 49, Hungary

ABSTRACT

Chirped dielectric rugate mirrors were constructed for broadband dispersion control in femtosecond laser oscillators by Fourier transform. Dispersive properties of the mirrors are tailored and explained on the basis of the time shifting theorem of Fourier analysis. Depending on their construction parameters, these chirped gradient-index structures exhibit high reflectivity and nearly constant negative, i.e. anomalous, group delay dispersion over frequency ranges well beyond the currently available fluorescence bandwidths of broadband laser-active materials. As a consequence, practical implementation of these novel dispersive devices would permit the full utilisation of the bandwidth offered by these broadband gain media, and allow the construction of small, compact femtosecond solid state laser oscillators generating optical pulses shorter than those achieved so far, directly from the laser oscillator.

1. INTRODUCTION

One of the main trends of laser physics today is the research and development of femtosecond laser sources\(^1\). The appearance of transition-metal-doped crystals having extremely broad fluorescence bands has brought great advances in this field allowing the construction of laser oscillators generating optical pulses of 10-100 fs directly at the laser output\(^1,2\). In the nineties, application of these femtosecond solid state lasers in the fields of physics, chemistry, biology, medicine and technology has made it possible to investigate temporal processes that could not be observed before due to the lack of measuring apparatus with high enough temporal resolution. There are two additional positive features of these systems originating from the short pulse durations: the low energy content and the high spectral bandwidth of every laser pulse. The former property significantly reduces the risk of damaging the samples, e.g. a semiconductor device being investigated by optical pulses, while the latter property allows us to apply them as broadband, high intensity, easy to handle light sources with very short emission times. Some of the most important applications of these ultrashort-pulse laser oscillators include the following: spectroscopic investigation of biological and chemical reactions, of semiconductors or other solid state materials with high temporal resolution\(^3\), investigation of strongly nonlinear optical processes, and high speed opto-electronic data transmission and processing systems. By utilising laser amplifier systems, the intensity of a single laser pulse can be boosted to the terawatt power range, which is widely used in plasma physics. In the long term, coherent X-ray sources (X-ray lasers) and new particle accelerators, that are smaller and cheaper than those used today, are planned to be built using these laser amplifier systems.

Femtosecond solid-state laser oscillators rely on a net negative, i.e. anomalous, intracavity group-delay dispersion (GDD) due to their soliton-like pulse shaping mechanism\(^1,2\). Until recently, prism pairs built in the laser cavity were the only low loss sources of broadband negative GDD. As a new physical concept, we have developed chirped multilayer high reflectors consisting of alternate discrete layers of high and low index dielectrics by using a standard computer optimisation technique\(^4\). The mirrors exhibited high reflectivity and nearly constant negative GDD of \(-45\) fs\(^2\) over a frequency range of 80 THz centred at 800 nm, and could be manufactured by means of a standard electron-beam evaporation technique\(^5\). To test these novel dispersive devices, a Kerr lens mode-locked Ti:sapphire laser was built, which consisted merely of cavity mirrors, a 2 mm-thick gain medium and an intracavity aperture. By taking advantage of several bounces on the dispersive mirrors in the cavity, the laser was able to generate optical pulses as short as 9 fs, which represents the best result reported to date\(^6\). Our experiments indicate that the
performance of mirror-dispersion-controlled (MDC) laser oscillators are likely to surpass their prism-pair controlled predecessors with some fundamental benefits, such as pulse quality (in terms of spectral symmetry and fitting the theoretical sech envelope), stability and compactness.

In this paper our purpose is to emphasise that more sophisticated optical coating design and deposition techniques may well lead to the practical implementation of extremely broadband dielectric high reflectors with significant, practically constant negative group delay dispersions. These special laser mirrors would allow us to utilise the full fluorescence band offered by broadband solid-state laser active materials, and generate optical pulses of a few optical cycles in duration. The dispersive mirror designs presented have been constructed by the use of Fourier transform, thereby resulting in chirped gradient-index dielectric structures. These chirped mirrors could be manufactured by the use of state of the art optical coating deposition techniques. Practical limitations to the performance of these dispersive devices are set by the index contrast and the total optical thickness of the dielectric coatings, both of which are basically determined by the coating materials available and the coating deposition technology used.

2. THEORY

The Fourier-transform technique is widely used for designing gradient-index optical filters, often termed as rugate filters, with prescribed spectral properties. All the work on this topic is based on the papers of Sossi and Kard, who showed that

\[ \frac{1}{2} \int_{-\infty}^{\infty} \frac{d \ln[n(x)]}{dx} \exp(ikx) dx = Q(k) \exp[i\Phi(k)] \]

(1)

where \( n(x) \) is the refractive index, \( k = 2\pi/\lambda \) is the wavenumber in air, and \( x \) is twice the optical distance from the centre of the inhomogeneous layer to physical position \( z \):

\[ x = 2 \int_{0}^{z} n(u) du \]

(2)

In Eq. (1), \( Q(k) \) is an appropriate function of the desired reflectance or transmittance. Using partial integration and a Fourier transform one can derive:

\[ \ln\left[ \frac{n(x)}{n_0} \right] = i \int_{-\infty}^{\infty} \frac{Q(k)}{k} \exp\left(i\left[\Phi(k) - kx\right]\right) dk \]

(3)

where \( n(\infty) = n (-\infty) = n_0 \), and \( Q(k) \) and \( \Phi(k) \) are even and odd functions of \( k \), respectively.

Design techniques based on Eq. (3) differ in the choice of \( Q(k) \) and \( \Phi(k) \), which are usually termed Q-function and phase factor, respectively. A variety of techniques exist because a simple, "universal" Q-function and phase factor has not been found; however, various approximate formulae have been successfully applied in the case of smooth refractive index dependence. It has been revealed that distortions in the Fourier-transformation originate from neglecting the multiple internal reflections in Eq. (1), see Refs. 23 and 24, the effect of which is striking in the case of dielectric structures consisting of discrete layers with high refractive index ratios \( n_i/n_j \). It has also been shown that from the physical point of view it could be more advantageous to view all optical thin film structures as reflection controlling devices. In ultrafast laser physics, it has been demonstrated that dynamic volume holograms
can be effectively utilized for ultrashort pulse shaping\textsuperscript{27}, and the reflection geometry is the optimal for this application. In Ref. 27 it has also been stated, that by controlling the amplitudes and phases of the gratings with various spatial frequencies forming the reflection hologram, one may control the amplitude and phase of each spectral component in a diffracted optical signal.

In the field of optical interference coatings, however, the phase factor for optical coating design has not been considered by earlier researchers due to the fact that phase change on reflection is rarely specified for these optical devices. Later on, it was recognized that solutions of rugate filter synthesis problems depend greatly on the choice of the phase factor\textsuperscript{20,22,25,26}. It has been shown that the phase shift on reflection is not uniquely connected with the amplitude reflectance modulus\textsuperscript{25}, thus it can be efficiently utilized to modify the refractive index profile without affecting the spectral performance. At first sight, it may seem to conflict with the well known Kramers-Kronig relation (KKR), which is often used to determine the refractive index of a bulk material based on intensity reflection \( R(k) \) measurements. However, in the case of inhomogeneous layers the KKR cannot be applied since reflectance depends basically on interference effects - a situation that is different from the classical case\textsuperscript{25}.

Our goal is to find a general formula which helps us in designing high performance dielectric high reflectors for dispersion control in femtosecond lasers, i.e. mirrors with prescribed dispersion properties. Bearing in mind the analogy between optical coatings and reflection holograms, and the results reported in Refs. 23 and 24, we choose the Q-function and phase factor as the amplitude and phase of the complex amplitude reflectance, \( r(k) \):

\[
Q(k) = |r(k)| \tag{4a}
\]

\[
\Phi(k) = \Psi'_r(k) \tag{4b}
\]

where \( \Psi'_r(k) = \arg [r(k)] \). We would mention, however, that from the mathematical point of view the ratio of the amplitude reflectance to the amplitude transmittance would be a much better function since this ratio has no singularities in the entire complex wave-number plane and also has a physical meaning\textsuperscript{25}. To simplify our treatment, we do not consider the frequency dependent phase shift for the transmitted electromagnetic field, since this has no practical importance in the case of dielectric high reflectors, when the transmittance is practically zero.

In spite of its approximate nature, Eq. (3) supplemented with Eqs. (4a) and (4b) is the crucial point of this paper. In the following, we demonstrate that the formula is well suited for constructing chirped dielectric rugate mirrors, which we regard as promising candidates for broadband dispersion controlling devices in femtosecond laser oscillators. Furthermore, these equations offer a deeper insight into the operation and mathematical description of chirped multilayer coatings built of discrete dielectric layers, which were designed for broadband dispersion control in femtosecond lasers\textsuperscript{4}.

In a number of papers\textsuperscript{20,24,26}, it has been shown that if one introduces a linear phase

\[
\Phi(k) = \Delta \chi k \tag{5}
\]

in Eq. (3), it results in a displacement \( \Delta \chi \) of the refractive index profile along the \( x \) axis. By differentiating with respect to \( k \), Eq. (5) can be rewritten in the form of \textsuperscript{26}

\[
\Delta \chi = \frac{d\Phi(k)}{dk} \tag{6}
\]

which is the time-shifting theorem of Fourier analysis. The theorem was successfully exploited to significantly reduce the optical thickness of synthesized rugate filters and control the shape of the refractive index variation without affecting the spectral performance\textsuperscript{20,21,26}. In Ref. 26, a general formula for the numerical calculation of the "optimal" phase function corresponding to a given design goal of
reflectance versus wavelength (or wavenumber) function has been presented; this formula results in thinner rugate filters or lower index contrast than alternatives that arbitrarily constrain the phase.

In connection with Eq. (6) it is worth noting that the frequency dependent group delay ($\tau$) upon reflection is calculated in a similar manner:

$$\tau = \frac{d\Psi_r(\omega)}{d\omega}$$

(7)

where $\Psi_r(\omega)$ is the frequency dependent phase change on reflection, and $\omega = ck$ is the angular frequency of the incident electromagnetic wave. In order to relate Eq. (6) to Eq. (7), we calculate the increase in the group delay ($\tau_{DP}$) upon reflection corresponding to the displacement of the refractive index profile, e.g. a dielectric mirror, simply by dividing the increase in the optical path $2\Delta x$ by $c$, the speed of light in vacuum:

$$\tau_{DP} = \frac{2\Delta x}{c}$$

(8)

Now we would like to comment on Eq. (3) in connection with gradient-index reflective structures, using some simple physical terms. First, non-zero reflectivity at wavenumber $k$ (or at wavelength $\lambda = 2\pi/k$) calls for a sinusoidal modulation in the logarithmic refractive index profile along the $x$ axis with a periodicity $\lambda_0 = \lambda/2$, corresponding to $k_0 = 2k$. The physics behind this claim is that if we have exclusively this spatial frequency, the infinitely large number of partially reflected beams originating from small Fresnel reflections within the structure meet in phase only for wavelength $\lambda$, whereas for other wavelengths, the vectorial sum of the reflected waves equals zero. Notice that the first term within the integral in Eq. (1), which can also be written in the form of $\frac{n'\prime(x)}{n(x)}$, stands for the reflectance amplitude due to Fresnel reflection at position $x$ inside the inhomogeneous dielectric layer. The expression describes the change in the index divided by the average index, which can be derived from the classical Fresnel formula. For higher reflectances, higher amplitude modulations, i.e. higher Fresnel reflections, are required, and the modulation for a given value of the amplitude reflectance is inversely proportional to the wavenumber $k$. Considering quarterwave stacks consisting of discrete layers, the latter property can be explained in the following way: if the mirrors had the same optical thickness but different periodicity, an increased number of reflective interfaces, i.e. a higher spatial frequency, would call for a smaller refractive index ratio ($n_2/n_1$) for the same peak reflectance, and vice versa. Secondly, supposing a Gaussian $\tau(k)$ amplitude reflectance spectrum, i.e. a Gaussian $\Phi(k)$ function with a phase factor set equal to zero $\Phi(k) = 0$, the extent of the refractive index profile along the $x$ axis obtained from the formula - more precisely: the half width of its Gaussian envelope is inversely proportional to the mirror bandwidth because of their Fourier-transform relationship. High mirror bandwidths with phase factors set to zero require high refractive index modulations over very short optical distances, a situation leading to physically unrealisable solutions.

In practice, the finite refractive index ratios set a limit to the bandwidth of the low dispersion reflective optics, such as high reflectors, dichroic mirrors and output couplers, which were developed for femtosecond laser oscillators containing prism pairs, and thus limit the ultimate bandwidth of these systems. This constraint on the cavity bandwidth could easily be overcome by the application of broadband (chirped) dielectric high reflectors. Another attractive feature of these mirrors is that they can provide a nearly constant negative GDD over most of the mirror bandwidth needed for solitary pulse formation in the cavity of a femtosecond solid-state laser oscillator.

In the following, chirped dielectric rugate mirrors are introduced by generalising the concept of shifted structures. The dispersive and spectral properties of multilayer rugate mirrors and chirped broadband rugate mirrors, synthesised by the use of Eq. (3), are calculated utilising the classic matrix multiplication technique and are then compared with their prescribed values.
3. CHIRPED BROADBAND RUGATE MIRROR WITH ANOMALOUS DISPERSION

3.1. Multiline, shifted rugate mirrors

To demonstrate the powerfulness of the Fourier-transform technique for designing dielectric mirrors with prescribed dispersion properties, first we consider the following mirror specification. We require high reflectivities at three separate wavelengths: \( \lambda_1, \lambda_2 \) and \( \lambda_3 \) corresponding to wavenumbers \( k_1, k_2 \) and \( k_3 \), respectively. The high reflectivity zones centred at these wavelengths should have a Gaussian shape with the same mirror bandwidth in terms of wavenumber, which is determined by parameter \( \sigma \). Furthermore, the group delay on reflection should be different at these separate wavelength regions in such a way that a wavepacket of the lowest carrier frequency corresponding to wavenumber \( k_1 = k_{01}/2 \) should suffer the largest group delay upon reflection, whereas a wavepacket of the highest carrier frequency, which corresponds to wavenumber \( k_3 = k_{03}/2 \), should have the smallest value. Based on our considerations presented in Section 2, we expect that the specification described above should be fulfilled by a multiline rugate mirror design obtained from Eq. (3) by using the following Q-function and phase factor, chosen as the amplitude and phase of the desired complex amplitude reflectance, \( r(k) \):

\[
Q(k) \exp(i\Phi(k)) = \sum_{i=1}^{3} \exp \left[ \frac{(k-k_{0i})^2}{2\sigma^2} \right] \exp[i\alpha_i(k-k_{0i})] \tag{9}
\]

The refractive index profile shown in Fig. 1a was generated by substituting \( Q(k) \) and \( \Phi(k) \) in Eq. (3) with the parameters: \( \sigma = 1.5 \, \mu m^{-1}, k_{01} = 2\pi / 0.4 \, \mu m^{-1}, k_{02} = 5/3 \, k_{01}, k_{03} = 7/3 \, k_{01}, a_1 = 4 \, \mu m, a_2 = 0 \, \mu m, a_3 = 4 \, \mu m. \) The refractive index of the surrounding medium has been set equal to \( n_s = 1.8 \). To achieve high enough reflectances, the modulation in the logarithmic refractive index profile has been increased by multiplying the right hand side of Eq. (3) by a factor of 35 in this specific case. Note, however, that it is nonphysical with respect to Eq. (4a), but the problem originates from the approximate nature of Eq. (3). Similarly, \( Q(k) \) may take values higher than 1 at some wavelengths in Eq. (9).

![Fig. 1a Refractive index profile of a multiline rugate mirror design described in the text](image)

As one would expect, the multiline rugate mirror design consists of three well defined substructures corresponding to the three separate reflection bands for wavenumbers \( k_{01}, k_{02} \) and \( k_{03} \). The substructures are shifted relative to the geometrical centre of the inhomogeneous layer as defined by Eq. (6), the time
shifting theorem of Fourier analysis. The corresponding shift parameters are $a_1 = 4 \, \mu m$, $a_2 = 0 \, \mu m$ and $a_3 = 4 \, \mu m$. It can also be clearly seen in the same figure that at lower frequencies a higher modulation in the refractive index profile is needed to provide the same reflectance and mirror bandwidth than at higher frequencies (as explained in Section 2).

By calculating the spectral performance of this multiline rugate mirror design, as shown in Fig. 1b along with the specified reflectance values, we find that the structure exhibits high reflectances only at $k_1 = k_{0d}/2 = 7.854 \, \mu m^{-1}$, $k_2 = k_{0d}/2 = 13.09 \, \mu m^{-1}$ and $k_3 = k_{0d}/2 = 18.176 \, \mu m^{-1}$, respectively, as required. Distortions in the shape of the reflectance bands originate from the multiplication factor we used in order to increase the refractive index modulation and the multiple internal reflections within the substructures, are neglected during Fourier transform.

![Graph showing reflectance vs. wavenumber](image1)

**Fig. 1b** Specified (dashed) and computed (continuous line) reflectance of multiline rugate mirror

With respect to the calculated group delay vs. wavenumber function (shown in Fig. 1c together with the specified values), we can conclude that the agreement between the specified and calculated values is fairly good. The group delay was computed numerically using the definition of Eq. (7).

![Graph showing group delay vs. wavenumber](image2)

**Fig. 1c** Specified (dashed) and computed (continuous line) group delay of multiline rugate mirror

It is mentioned that a constant group delay of $\tau_0 = 2 \times 6.388 \, \mu m^2 / (0.3 \times 10^{-9} \, m/s) = 42.583 \, fs$ has been added to the calculated values. This group delay corresponds to the physical position mismatch between the centre of our inhomogeneous layer, which is our $x = 0$ position during the Fourier transform, and the
top of the layer at the position of $x = 6.388 \ \mu m$, where the phase shift on reflection is calculated. The slight parabolic change in the group delay functions around the mirror central frequencies is similar to the behaviour of quarterwave stacks consisting of discrete layers. It originates from the increasing penetration depth of the electromagnetic field when the detuning from the mirror central frequency increases, as has been explained in detail elsewhere\textsuperscript{6,30}. This behaviour results in a positive third-order dispersion (TOD) of the mirrors, which is calculated as the third derivative of the phase change on reflection $\Psi_r$ with respect to the angular frequency $\omega$.

With respect to multinline, shifted rugate mirrors we can conclude that a frequency dependent group delay function can be tailored by placing mirrors of different periodicities and finite bandwidths at different positions determined by Eqs. (5-8). Generalising this approach, we now introduce the concept of chirped dielectric rugate mirrors in which the period of the refractive index profile is linearly varied along the $x$ axis in order to obtain high reflectivity and an approximately linearly decreasing group delay vs. frequency (or wavenumber) function of over broad bandwidths.

### 3.2. Chirped rugate mirrors

We define a second order phase factor $\Phi(k)$ as the function wavenumber, i.e. a second order phase shift on reflection (see Eq. (3b)), written in the following form:

$$\Phi(k) = d_0 + d_1 (k - k_0) + d_2 (k - k_0)^2$$  \hspace{1cm} (10)

Furthermore, we define the Q-function $Q(k)$, i.e. the modulus of the complex amplitude reflectance (see Eq. (3a)), as a Gaussian function:

$$Q(k) = \exp \left[ -\frac{(k - k_0)^2}{2\sigma^2} \right]$$  \hspace{1cm} (11)

Calculating the frequency dependent displacement $\Delta x(k)$ of the refractive profile along the $x$ axis using Eq. (6) one obtains:

$$\Delta x(k) = d_1 + 2d_2 (k - k_0)$$  \hspace{1cm} (12)

or equivalently, using Eq. (8):

$$\tau_{DP}(k) = \frac{2d_1 + 2d_2 (k - k_0)}{c}$$  \hspace{1cm} (13)

which is a linear function of the wavenumber, thus the angular frequency of the incident electromagnetic field.

Equation (12) shows that the different spatial frequency components are shifted linearly along the $x$ axis as a function of $k$, i.e. the second-order phase term in Eq. (10) results in a chirped dielectric rugate structure (see Fig. 2a). The parameters that we used during its computations using Eq. (3) were: $k_0 = 2\pi/0.4 \ \mu m^{-1}$, which corresponds to our selected mirror central wavelength of 0.8 $\mu m$, $\sigma = 1.4 \ \mu m^{-1}$, $d_1 = 0 \ rad$, $d_2 = 0 \ \mu m$, $d_3 = 0.589 \ \mu m^2$. The refractive index of the surrounding medium has been set equal to $n_0 = 1.8$, as in the case of the multinline rugate mirror discussed earlier. To obtain high enough reflectances, we multiplied the right hand side of Eq. (3) by a factor of 8, however this led to a practically unrealisable solution. Combination of the optimization formula presented in Ref. 26 and the concept of
chirped dielectric rugate mirrors should result in practical, broadband chirped rugate mirror designs with anomalous group delay dispersions.

![Refractive index profile of a chirped rugate mirror design described in the text](image)

**Fig. 2a** Refractive index profile of a chirped rugate mirror design described in the text

In Fig. 2b, the computed reflectance of the chirped structure is presented along with the prescribed reflectance values. It is worth noting that the structure exhibits practically 100% reflectance from wavenumbers of 6.35 μm⁻¹ to 9.35 μm⁻¹, which correspond to wavelengths of 0.989 μm, and 0.671 μm, respectively.

![Specified (dashed) and computed (continuous line) reflectance of chirped rugate mirror](image)

**Fig. 2b** Specified (dashed) and computed (continuous line) reflectance of chirped rugate mirror

Calculating the group delay introduced by the chirped mirror by using Eq. (7), the result of which is shown in Fig. 2c, we have found that the group delay decreases monotonously as the function frequency (or wavenumber) over most of the high reflectivity band of the mirror. The mirror exhibits nearly constant GDD over the 6.5-9 μm⁻¹ wavenumber range corresponding to the wavelength range of 0.966 - 0.698 μm. It is again mentioned, that a constant group delay of \( \tau_0 = 2 \times 6.388 \, \mu \text{m} / (0.3 \times 10^{-9} \, \text{m/s}) = 42.583 \, \text{fs} \) has been added to the prescribed group delay values as explained in connection with Fig. 1c. Despite the calculated group delay vs. wavenumber function of the chirped structure apparently differing from the specified linear function, the design could certainly be efficiently utilised in femtosecond solid state laser oscillators.
Fig. 2c  Specified (dashed) and computed (continuous line) group delay of chirped rugate mirror

4. CONCLUSIONS

The concept of chirped rugate mirrors with anomalous group delay dispersions has been introduced. Because of their high reflectivity and nearly constant negative GDD over extremely broad bandwidths, we regard them as promising candidates for broadband dispersion control in femtosecond laser oscillators. The mirror designs presented have been constructed for illustration only, and further theoretical work is needed to adapt them to different coating deposition techniques and evaporation systems.

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6. REFERENCES


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