Chirped multilayer coatings for broadband dispersion control in femtosecond lasers

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Optical thin-film structures exhibiting high reflectivity and a nearly constant negative group-delay dispersion over frequency ranges as broad as 80 THz are presented. This attractive combination makes these coatings well suited for intracavity dispersion control in broadband femtosecond solid-state lasers. We address design issues and the principle of operation of these novel devices.

The relevance of intracavity dispersion control in passively mode-locked ultrashort-pulse laser was recognized soon after the appearance of the first systems operating in the femtosecond domain.[1] Negative dispersion that is due to wavelength-dependent refraction in a pair of Brewster-angled prisms combined with positive material dispersion proved to be an efficient and convenient means of controlling the net group-delay dispersion (GDD) inside the laser cavity.[2]

In solid-state lasers femtosecond pulse generation invariably relies on a net negative intracavity GDD because of an ultrafast self-phase modulation caused by the optical Kerr effect in the laser medium. Hence prism pairs have become standard components in these systems. The interplay between negative GDD and Kerr-induced self-phase modulation, often referred to as solitonlike shaping, appears to be the dominant pulse-forming mechanism that determines the steady-state pulse duration in femtosecond solid-state lasers.[3] In practical prism-pair-controlled broadband laser systems a major limitation to ultrashort pulse generation originates from the variation of the intracavity GDD with wavelength. The principal source of this high-order dispersion was found to be the prism pair.[3,4] In this Letter we report the novel development of chirped multilayer mirror coatings that can exhibit essentially constant negative GDD over a frequency range as broad as 80 THz. Careful design permits higher-order contributions to the mirror phase dispersion to be kept at low values or to be chosen such that high-order phase errors introduced by other cavity components (e.g., the gain medium) are canceled. Replacing the prism pair with these novel devices offers the potential of generating pulses that are shorter than previously achievable directly from the laser. In addition, this simplifies the cavity design and may permit the construction of more compact and reliable femtosecond sources.

The first thorough investigations of the frequency-dependent phase retardation (phase dispersion) of multilayer dielectric coatings date back to the early 1960's.[5,6] The emergence of femtosecond lasers in the 1980's has led to a revival of interest in this field.[7–13] Whereas standard quarter-wave dielectric mirrors were shown to introduce negligible dispersion at the center of their reflectivity bands,[7,8,11] various specific high-reflectivity coatings (Gires–Tournois interferometers, double-stack mirrors, etc.) with adjustable GDD (through angle tuning) were devised and used for the precise control of intracavity dispersion in femtosecond dye lasers.[14,15] However, the GDD introduced by these mirror coatings is accompanied by high cubic and higher-order dispersive contributions. As a consequence, a constant GDD could be obtained only over a limited wavelength range (<10 THz). The difficulty in realizing broadband GDD control relates to the physical origin of dispersion in these devices; different frequency components are trapped for different periods of time in Fabry–Perot-like resonant structures.

A distinctly different type of GDD arises from the wavelength dependence of the penetration depth of the incident optical field in multilayer coatings. This effect does not rely on the presence of resonant structures and offers the possibility of realizing a GDD that is a slowly varying function of wavelength over a broad bandwidth. A constant GDD requires a group delay that varies approximately linearly with the wavelength. A wave packet of a given center wavelength is most efficiently reflected by a corresponding quarter-wave stack. Therefore a monotonic variation of the multilayer period throughout the deposition process (chirped coating) should result in a penetration depth[16] (and thus group delay) that varies monotonically with the wavelength. However, a previous study of chirped multilayer coatings with layer thicknesses following monotonic variations revealed that the GDD is strongly perturbed by some Fabry–Perot-like resonances in these simple structures.[17] Our studies have indicated that the undesirable resonant features can be almost completely eliminated by

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slight adjustment of the layer thicknesses. This finding has been a most important step toward the practical realization of dispersive mirrors having a group delay that is a linear function of optical frequency. In addition, chirped multilayer coatings have the potential for extending the bandwidth of standard low-dispersion quarter-wave mirrors.

A preliminary design utilized a structure consisting of 42 alternating layers of SiO_2 ($n = 1.45$) and TiO_2 ($n = 2.3$) with optical thicknesses close to a quarter of 0.8 μm, our selected center wavelength. The multilayer period was slightly increased near the substrate and decreased at the air–coating interface to produce a group delay that increased with wavelength, i.e., negative GDD. The mirrors were required to be highly reflecting and have a constant negative GDD over the wavelength range 710–900 nm. Alternatively the GDD could be required to exhibit a slight linear variation with a slope suitable for compensating the cubic phase dispersion of the gain medium. This is in strong contrast to prism pairs, in which the ratio of cubic to quadratic dispersion is determined by material parameters and the operation wavelength.[18] A simple computer refinement algorithm[19] was used to minimize the quadratic deviation of the complex reflectivity-versus-frequency function of the actual mirror design from the required specification. The quadratic error was made up of two parts, for the amplitude and the phase characteristics, whose relative weights could be independently adjusted. The prescribed value of negative GDD was increased stepwise until the quadratic error started to increase rapidly. Accordingly the operating negative GDD was found to trade off against a high reflectivity and a low variation of the prescribed GDD-versus-wavelength function.

This optimization procedure enabled us to realize the dispersive mirror design shown in Fig. 1 (Ref. 20) by use of a standard electron-beam evaporation technique.[13] Our obtained structure preserved the feature of increasing layer period toward the substrate, even though the variation is far from what we consider as linear. The calculated reflectivity of this design is larger than 99.9% around the center wavelength and drops to ≈99.5% at 710 and 900 nm. The nominal GDD of the mirror is ≈ -45 fs^2 at $\lambda = 800$ nm, with a slight linear variation yielding a cubic dispersion of ≈ -33 fs^3, which permits a simultaneous compensation of GDD and cubic dispersion in a Ti:sapphire laser.

The wavelength dependence of the GDD exhibits a weak oscillatory behavior, with deviations less than ± 5 fs^2 from the required (linear) GDD function over the wavelength range of 720–890 nm (≈80 THz). The GDD of -45 fs^2 corresponds to a group-delay difference of ≈22 fs between the extremes of this spectral range. As we show below, for a 42-layer design this is close to the maximum delay difference that can be attained without making use of resonances. Any further increase in GDD could be achieved only at the expense of resonant structures (e.g., Gires–Tournois interferometers) appearing in the multilayer design, which implies large variations in the GDD over the mirror reflectivity range.

Figure 2 plots the computed electric-field distribution inside the dielectric mirror as a function of wavelength. As required, the penetration depth (and thus the group delay) increases approximately linearly with the wavelength. The figure also gives clear evidence of the high reflectivity of the mirror between 700 and 900 nm, as indicated by the disappearance of the optical field at the substrate–coating interface (optical distance 0).

The computed group delay (first derivative of the reflected phase with respect to frequency) as a function of wavelength is depicted in Fig. 3 along with measured data. The experimental data were obtained by use of the white-light interferometer technique of Knox *et al.*[11] To reduce the experimental errors, we used 16 reflections at a ≈10° angle of incidence in the interferometer, resulting in a measurement accuracy of ≈1 fs. These results demonstrate that dielectric mirrors with approximately constant GDD over a broad spectral range can be designed and fabricated by use of standard computation and evaporation techniques, respectively.

The maximum achievable negative GDD is limited by the maximum group-delay difference that can be obtained between the extremes of the reflectivity range. This in turn relates to the optical thickness

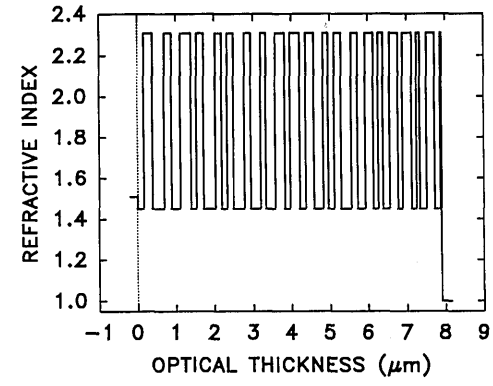

Fig. 1. Theoretical refractive-index profile of a high-reflectivity TiO_2–SiO_2 multilayer coating designed specifically for broadband GDD control in femtosecond lasers.

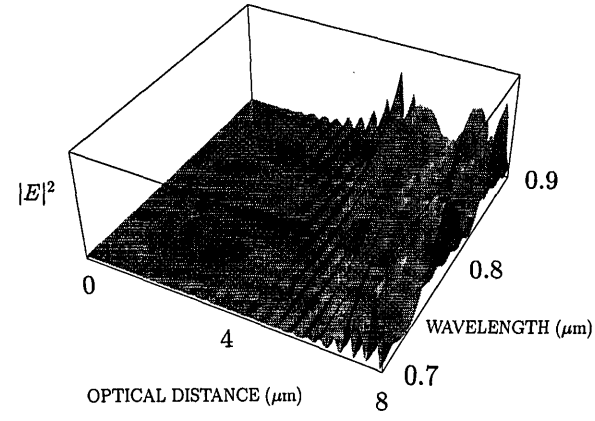

Fig. 2. Computed electric-field distribution as a function of wavelength in the chirped dielectric structure shown in Fig. 1.

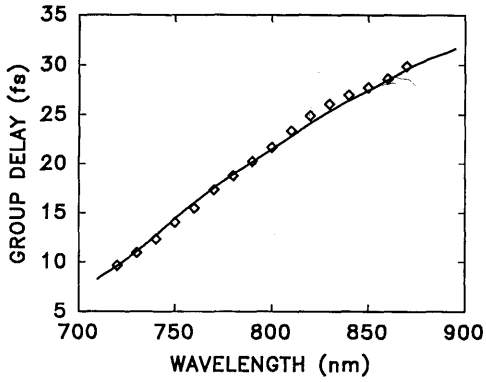

Fig. 3. Computed group delay as a function of wavelength (solid curve) together with experimental data (squares) for the multilayer design of Fig. 1. Note that the absolute delay could not be measured; therefore a wavelength-independent constant delay was added to the measured relative data.

of the coating. We have found that a simple approximate expression for the maximum achievable group-delay difference can be written as

$$\Delta\tau_{\max} = \frac{2(t_{\text{chirped}} - t_{\text{qw}})}{c}, \tag{1}$$

where t_{chirped} is the optical thickness of the chirped multilayer coating and t_{qw} is that of a standard quarter-wave high reflector ($R > 99.9\%$) consisting of the same pair of alternating layer materials. In physical terms, the required high reflectivity of the dispersive mirror calls for a minimum optical thickness of $t \approx t_{\text{qw}}$, and only excess layers can introduce an appreciable frequency-dependent group delay around the center of the high reflectivity band. Assuming that the group delay varies in a linear manner with frequency, we see that the corresponding upper estimate for the GDD is given simply by the ratio of $\Delta\tau_{\max}$ to the mirror bandwidth $\Delta\omega$.

For the specific case of TiO_2–SiO_2 mirrors centered around $\lambda \approx 0.8$ μm we have $t_{\text{qw}} \approx 4$ μm, yielding $\Delta\tau_{\max} \approx 27$ fs for our 8-μm-thick structure, in reasonable agreement with the results presented in Fig. 3. With the number of layers fixed, $\Delta\tau_{\max}$ scales linearly with the chosen center wavelength of the dispersive mirror. For a selected operating wavelength, we can increase $\Delta\tau_{\max}$ and thus the magnitude of broadband negative GDD only by increasing the number of layers, which is limited by scattering and absorption losses that are due to structural defects and impurities in the deposited layers, respectively.[13] It is expected that more sophisticated coating techniques will permit the production of higher-quality layers and thereby open the way toward the realization of more complex structures with higher values of the negative GDD over bandwidths approaching 100 THz.

In summary, we have reported a novel dielectric mirror providing approximately constant negative dispersion over a bandwidth as broad as 80 THz. The presented mirror design can be adopted for any broadband mode-locked solid-state laser operating in the wavelength range of 0.5–2 μm by a simple rescaling of the layer thicknesses. With suitable layer materials this spectral range can be extended well into the ultraviolet and infrared spectra. Furthermore, engineering the wavelength dependence of the penetration depth might lead to interesting applications in other areas of physics in which scattering and interference in quasi-periodic structures take place.

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