Relationships among group delay, energy storage, and loss in dispersive dielectric mirrors

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We show that absorbed and stored electromagnetic energy are proportional to the reflection group delay in highly reflective dispersive dielectric mirrors over the high-reflectivity band. Our theoretical considerations are verified by numerical simulations performed on different dielectric mirror structures. The revealed proportionality between group delay and absorbed energy sets constraint on the application of ultrabroadband and/or dispersive dielectric mirrors in broadband or widely tunable, high-power laser systems.

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In the last two decades, theoretical and experimental investigations on reflection delay time of optical pulses reflected by multilayer dielectric structures have been of scientific interest because frequency-dependent group delay (GD) of the dielectric mirrors can be well suited for intracavity or extracavity dispersion compensation of ultrashort pulse lasers[1]. Aperiodic dielectric mirror structures, which are often referred to as chirped mirrors, are advantageous, not only because they introduce a certain amount of negative dispersion, but also because they exhibit a considerably broader high-reflectance band than standard, low-dispersion quarter-wave (QW) mirror stacks. Aside from chirped mirrors, there is another group of dispersive mirrors referred to as: Gires-Tournois interferometer-type mirrors[2]. The fact that a relationship exists between the electromagnetic energy stored in the volume of a thin-film structure and its GD is known in the field of telecommunication technology[3]. This relationship is also a starting point in resolving the long-haul scientific problem of superluminal delay times of electromagnetic wave packets during transmission through highly reflective, lossless photonic bandgap structures[4]. In practical laser systems, however, one usually cannot neglect the absorption (or scattering) loss of dielectric multilayer mirrors because laser performance, such as intracavity loss, beam quality, maximum output power, laser damage threshold power, and others, strongly depends on these physical quantities. From this aspect, one can address the question of whether a general relationship exists between the reflection GD and the absorption loss of a dielectric multilayer mirror (throughout the letter, we mean one-photon or linear absorption when we use the word “absorption”). For QW-stack mirrors, GD and absorptance have been shown as proportional to each other at the central wavelength of the QW mirror when the loss is sufficiently low[5]. For a specific multistack multilayer mirror design, Ferencz *et al.* also found conspicuous, but unexplained, proportionality of these two physical quantities[6]. For a nonspecific, general, highly reflective dielectric mirror structure, however, neither the relationship between GD and the absorptance nor the relationship between GD and the stored energy in the presence of loss has been investigated systematically thus far.

In this letter, we first theoretically deduce the relationship among group delay, stored energy, and absorptance of a highly reflective multilayer mirror of a general design, after which we verify our theoretical results by numerical simulations. Proportionality between the absorptance and the GD is very important for application of ultrabroadband chirped mirrors (UBCMs)[7] or highly dispersive mirrors[8] in high power, pulsed laser systems: these mirrors are used for broadband feedback and dispersion compensation, and their reflection GD is typically one or two orders of magnitude higher than that of the low-dispersion QW stacks. However, low reflection loss is crucial in case of dielectric mirrors designed for such lasers: the absorbed power of the laser beam is transformed to heat, which can lead to wavefront distortion or, worse, damage to the coating due to a linear absorption process, which is the dominating damage mechanism for >50 ps pulse durations[9]. For shorter pulses, one should also consider nonlinear processes, such as two-photon absorption or avalanche ionization, which are beyond the scope of this letter. Dielectric mirrors exhibiting relatively high GD on reflection, however, are not exclusively used in ultrashort pulse laser systems. UBCM structures[7] are widely used in broadly tunable CW or ns pulse lasers or in optical parametric oscillators and amplifiers for beam steering or feedback over a wide frequency range. In these applications, linear absorption loss, subsequent heating, and mirror damage could be critical issues. Loss in dielectric mirrors becomes higher and a more important issue as we move to shorter wavelengths, especially in the UV spectral range.

As a starting point in our theoretical investigations, we recall the main results in Ref. [4], in which the transmission of a relatively narrowband light pulse through a lossless photonic barrier was investigated. Using the continuity equation for energy, Ref. [4] analytically showed that the weighted sum of the reflection GD (τ_{gr}) and the transmission GD (τ_{gt}) is equal to the so-called dwell time (τ_{d}), which is the ratio of the stored energy (U) and the incident power (P_{i}):

$$R\tau_{\mathrm{gr}} + T\tau_{\mathrm{gt}} = \frac{U}{P_{\mathrm{i}}} \equiv \tau_{\mathrm{d}}, \qquad (1)$$

where $R = |r|^2$ and $T \propto |t|^2$ are the power reflectance and transmittance, respectively, and $r(\omega)=|r(\omega)|\exp[\mathrm{i}\phi_\mathrm{r}(\omega)]$ and $t(\omega)=|t(\omega)|\exp[\mathrm{i}\phi_\mathrm{t}(\omega)]$ are the complex amplitude reflection and transmission coefficients, respectively. The GDs, τ_gr and τ_gt, are the first derivatives of the phase shifts, $\phi_\mathrm{r}(\omega)$ and $\phi_\mathrm{t}(\omega)$, with respect to the angular frequency (ω), respectively. In the cited work, the bandwidth of the pulse was assumed to be so narrow that the magnitudes of t and r are constant over the laser spectrum and that τ_gr and τ_gt are much smaller than the pulse duration. The study also showed that, for a lossless barrier, dwell time equals 1/e lifetime of stored energy in the barrier, that is, the cavity lifetime, τ_c[4]:

$$\tau_\mathrm{c} \equiv \frac{Q}{\omega} = \tau_\mathrm{d}, \tag{2}$$

where Q represents cavity quality factor.

Let us now assume that the barrier is a dielectric mirror with high reflectance ($R \approx 1$) over a specific bandwidth. Therefore, the $T\tau_\mathrm{gt} \ll R\tau_\mathrm{gr}$ relation holds within the mirror bandwidth in the absence of very strong Fabry-Perot-like resonances, causing extremely high τ_gt values. In this case, according to Eq. (1), $\tau_\mathrm{gr} \approx U/P_\mathrm{i}$; thus, the reflection GD (simply called the GD when mirrors are considered) is approximately proportional to the stored energy if the incident power is constant:

$$\tau_\mathrm{gr} \propto U. \tag{3}$$

If the barrier has some energy dissipation, the expectations used in the derivation of Eqs. (1) and (2) are no longer precisely valid. However, high-quality dielectric multilayer mirrors grown by state-of-the-art deposition techniques, such as ion-assisted deposition or ion beam sputtering have very low loss. Therefore, the $A \ll R$ condition applies, where A denotes the absorptance of the mirror. In this case, one can expect that Eqs. (1) and (2) provide a good approximation. From Eq. (2) and from $\tau_\mathrm{gr} \approx U/P_\mathrm{i}$, we obtain $\tau_\mathrm{c} \approx \tau_\mathrm{gr}$. Therefore, a higher GD accompanies a longer decay time of the stored energy (i.e., with a longer "cavity" lifetime of a photon) in the mirror's volume. Because the probability of an absorption event scales with the cavity lifetime, one can expect that the absorptance of a slightly absorbing dielectric mirror is proportional to the GD as well (at a certain wavelength), assuming that the different layers of the mirror have a uniform loss:

$$A \propto \tau_\mathrm{gr}. \tag{4}$$

Next, we check the validity of Eqs. (3) and (4) by numerical calculations performed on different types of mirrors. We address the question how much these relations are affected by the loss of the layer materials. All the calculations used the transfer-matrix method, commonly used in thin-film physics, for normal incidence of light. The four mirror structures we examined are a QW stack, an UBCM, and two multicavity Gires-Tournois interferometer (MCGTI) mirrors denoted by HS MCGTI and LS MCGTI, all with a central wavelength of 800 nm. The QW stack is described by S | (H L)^{10} H | A, where S, H, L, and A denote the substrate, the high- and the low-refractive-index layers of quarterwave optical thickness, and air, respectively. The UBCM design is the same as listed in Ref. [7], whereas the HS MCGTI design is the reoptimized version of a structure reported in Ref. [8]. We designed the LS MCGTI to have nearly the same overall optical thickness as the HS MCGTI mirror and a very similar GD versus wavelength function. The refractive index profiles of the MCGTI mirrors are depicted in Fig. 1 (thin black lines). The computed GD versus wavelength functions along with the absorptance versus wavelength functions of the four mirror structures are shown in Fig. 2, where the extinction coefficients of the low- and high-index layers are set to $k_\mathrm{L}=10^{-5}$ and $k_\mathrm{H}=10^{-4}$, respectively.

The GD of the QW mirror and the UBCM are basically determined by the frequency-dependent penetration depth of the electromagnetic field[1−3]. On the contrary, cavity-like, resonant "spacer" layers exist in the MCGTI structures (the four thicker layers in Fig. 1), in which a high standing wave field builds up at different wavelengths, causing extremely high GD values[2,3,8]. Three of the four spacer layers of the HS MCGTI structure are made of the higher-index material. On the contrary, all the four spacer layers of the LS MCGTI mirror are made of the lower-index material. Low-index coating materials typically have a lower absorption; therefore, we expect a lower reflection loss in the LS MCGTI structure because most of the standing-wave electric field is supposed to be confined in the spacer layers. However, after the numerical calculations, we find that the absorptance versus wavelength functions of the two MCGTI mirrors do not differ significantly. This finding can be explained by the fact that a significant amount of electromagnetic energy is distributed and stored all over the mirror structure, as shown in Fig. 1.

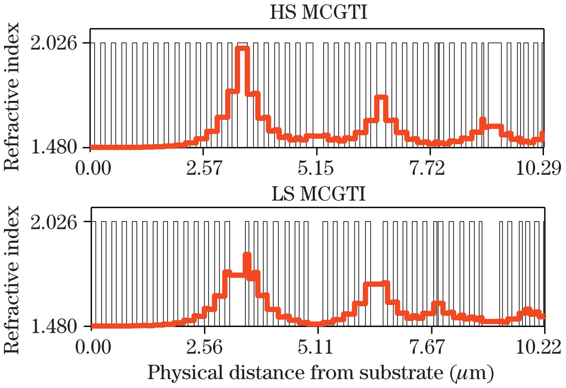

Fig. 1. (Color online) Computed electromagnetic energy density distributions (thick red lines) inside the two MCGTI multilayer structures at the peak resonance wavelength of 830 nm.

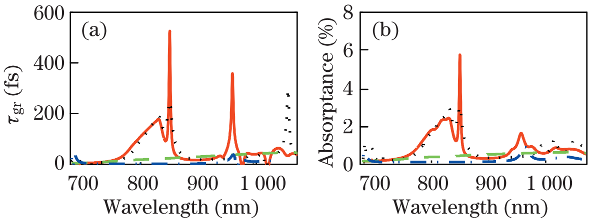

Fig. 2. (Color online) (a) GD and (b) absorptance versus wavelength functions of the QW stack (dash-dotted blue line), the UBCM (dashed green line), the HS MCGTI mirror (dotted black line), and the LS MCGTI mirror (solid red line).

We then investigate the question how the amount of dissipated power in the mirrors affects the proportionality between the GD and the stored energy described by Eq. (3), and between the GD and the absorptance described by Eq. (4), respectively. This analysis is carried out by first plotting the stored energy or the absorptance versus the GD functions, then fitting a straight line with zero intercept on the datasets, and, finally, characterizing the quality of the fit with the adjusted R-square parameter[10]. We also perform general linear fits on the curves, with a not-necessarily-zero intercept, and determine the degree of correlation between the examined quantities this way. The analysis can be conducted with two different approaches: one can take a given mirror and investigate Eqs. (3) and (4) considering different wavelengths within a certain wavelength range, or one can take several mirrors made from the same materials and having different GDs on a given wavelength. In both cases, the analysis is performed within the highly reflecting, and/or dispersive band of the mirrors, which are referred to as the "usable band" of the mirrors throughout this letter. For the QW stack, this band is defined by a required minimum value of reflectivity ($R>99.748\%$ for the lossless case), resulting in the 715–909 nm range. The dispersive mirrors (the UBCM and the two MCGTI mirrors) are typically used in the spectral range, where their dispersion is nearly constant within the high-reflectance band. For this reason, the usable band of the UBCM is defined as the 720–1 050 nm range (where $R>99.748\%$ for the lossless case), and this band of both MCGTI mirrors is defined as the 780–825 nm range, where $R>99.69\%$ for the HS MCGTI mirror and $R>99.30\%$ for the LS MCGTI mirror, also for the lossless case. Within these spectral ranges, no such resonances exists wherein the condition of $T\tau_{\mathrm{gt}} \ll R\tau_{\mathrm{gr}}$ would be impaired. Regarding each mirror structure, ten cases having different layer loss parameters were investigated. The corresponding extinction coefficients (k) of the layer materials are listed in Table 1. The sum is $k_{\mathrm{L}}+k_{\mathrm{H}}$; thus, the total loss increases for increasing column numbers.

First, we analyzed each mirror within its usable band separately. To make the analysis straightforward, all of the physical parameters used in the calculations were considered wavelength independent (i.e., the incident power spectral density (P_{i}), the imaginary part of the complex refractive indices (i.e., the extinction coefficients, k) and also their real parts (n) for each mirror layer). The (wavelength-dependent) absorptance and the stored energy spectral density were plotted against the (also wavelength dependent) GD. The best linear fits having arbitrary or enforced zero intercept were determined for each extinction coefficient combination listed in Table 1. The goodness of each fit is characterized by the deviation of the adjusted R-square parameter value from one: DAR≡1–Adj.R-square. The smaller the DAR value, the better the fit. The results are shown in Fig. 3.

Table 1. Extinction Coefficients of the Layer Materials

Serial No.	1	2	3	4	5	6	7	8	9	10
$k_{\mathrm{L}}(\times 10^{-5})$	0	1	1	5	5	10	10	50	50	100
$k_{\mathrm{H}}(\times 10^{-5})$	0	1	5	5	10	10	50	50	100	100

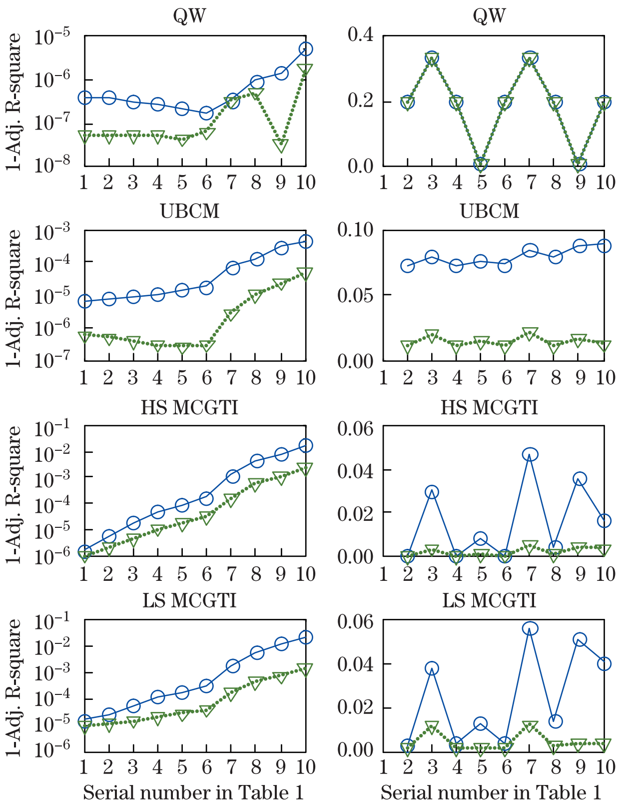

Fig. 3. DAR values obtained for the stored energy versus GD functions (left side, on log scale) and for the absorptance versus GD functions (right side) of the four mirrors, each mirror analyzed separately within its usable band. Triangles: general linear fits; circles: linear fits with enforced zero intercept.

Regarding the stored energy versus GD datasets, a linear fit with zero intercept can provide a considerably good fit, even if the extinction coefficients are as high as 10^{-3}, which is much higher than typical k values of laser-grade optical-coating materials in the UV, VIS, and NIR range. A general linear fit, which does not result in a zero intercept by necessity, gives even better results. A higher loss generally results in a higher DAR value (i.e., the degree of correlation between the stored energy and the GD reduces); however, the fit is still very good. Referring to the linear fits on the absorptance versus GD datasets, we can say that the fits are also quite good, except for the QW mirror. From the practical point of view, however, the absorptance versus GD relation is much more critical for dispersive mirrors, for which the correlation between these two physical quantities are still quite good. The goodness of fit on the absorptance versus GD datasets rather depends on the $k_{\mathrm{H}}/k_{\mathrm{L}}$ ratio than on the total mirror loss.

After this analysis, we must note that the actual loss of a layer is better determined by the absorption coefficient, $\alpha=4\pi k/\lambda$ (where λ is the vacuum wavelength), than by the extinction coefficient (k) we have used thus far. Therefore, even if k is wavelength-independent, the loss is determined by a wavelength-dependent parameter, which can influence our analysis regarding the relation between absorptance and GD. Therefore, we performed similar calculations on mirrors composed of the same optical materials, but of different structures, at a certain wavelength. The new mirror structures were obtained

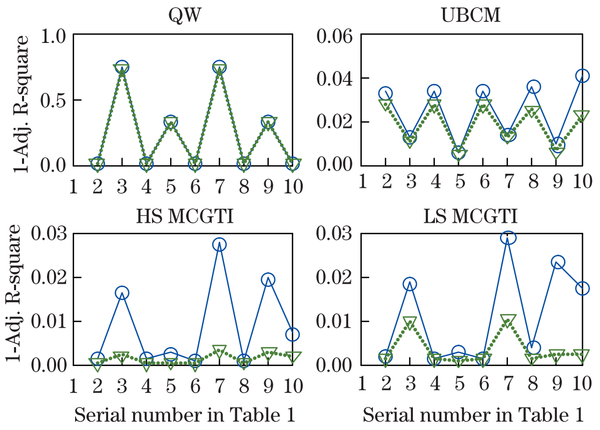

Fig. 4. DAR values for the absorptance versus GD datasets obtained at a certain wavelength for different mirror structures of rescaled physical thickness values of the layers. Triangles: general linear fits; circles: linear fits with enforced zero intercept.

by rescaling the physical thickness values of each of the four different mirror structures we have studied thus far. For these new structures, the DAR values on the absorptance versus GD datasets are depicted in Fig. 4. For the UBCM, the new values of the DAR parameter have become much smaller than in our previous analysis, where α had a considerable change over the relatively broad high-reflectance band. For the MCGTI mirrors, however, the new results are very similar to our earlier results due to their relatively narrow usable bandwidth (45 nm). The QW stack has an anomalous behavior: the DAR values are even higher than before for the odd serial numbers (i.e., where $k_{\mathrm{H}} \neq k_{\mathrm{L}}$).

In conclusion, our results show that, in dispersive dielectric multilayer mirrors, the stored energy as well as the absorbed power is well proportional to the reflection GD of the mirrors within their usable high-reflection band. The proportionality between the absorption loss and the GD has several practical consequences, especially regarding ultrabroadband dielectric mirrors (which are inherently dispersive) when they are used in broadband or broadly tunable, high-power laser systems. In short, care should be taken to minimize the absorption loss and the consequent detrimental thermal effects. First, when designing such mirrors, both the offset value of GD and the GD dispersion, should be kept as low as possible. For minimum dispersion, one should use high-refractive-index difference. This latter requirement is a tradeoff with the need for low-extinction coefficients of the layers because higher refractive index materials generally have higher k values. Additionally, the mirrors should always have a negative dispersion (i.e., an increasing GD towards longer wavelengths) because both the absorption and the scattering loss of layer materials typically increase towards shorter wavelengths. In addition to these design criteria, such mirrors should be manufactured by state-of-the-art deposition techniques.

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