Femtosecond time-resolved reflectometry measurements of multiple-layer dielectric mirrors

A. M. Weiner,* J. G. Fujimoto, and E. P. Ippen

Department of Electrical Engineering and Computer Science and Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

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We describe the use of optical pulses as short as 16 fsec for time-resolved reflectometry studies of multiple-layer dielectric mirror coatings. Pronounced pulse-distortion effects have been observed following reflection from broadband dielectric mirrors.

Recent theoretical investigations indicate that dispersion arising from multiple-layer dielectric mirror coatings may produce significant pulse-shaping effects in the femtosecond regime.[1] An understanding of these processes is especially relevant to the design and operation of femtosecond lasers. For example, the pulse widths of colliding-pulse mode-locked (CPM) ring dye lasers[2] are particularly sensitive to the ensemble of mirrors in the cavity.[3,4] We have recently generated pulses as short as 16 fsec by compression in an optical fiber.[5,6] In this Letter we describe the application of these compressed pulses to perform time-domain reflectometry measurements on multiple-layer broadband dielectric mirrors. The compressed pulses permit the observation of pronounced pulse-shaping effects that would not be evident with longer pulses.

A detailed description of the procedure for the generation and measurement of pulses as short as 16 fsec has been reported.[5,6] The femtosecond laser source for our pulse-compression experiments consists of a CPM ring dye laser[2] and a multiple-stage, high-power, femtosecond dye amplifier.[7] The system produces 65-fsec pulses at approximately 625 nm with a repetition rate of 10 Hz. With about 5 nJ of energy coupled into the short, 8-mm length of single-mode optical fiber, the input spectrum broadens by a factor of 4 because of self-phase modulation. The actual compression to 16 fsec is accomplished using a pair of diffraction gratings, which also precompensates for dispersion in the measurement apparatus.

Pulse-duration measurements are performed by autocorrelation using noncollinear second-harmonic generation in a KDP crystal.[8] As discussed previously,[5,6] special precautions must be taken to avoid instrumental limitations arising from geometrical and dispersive effects in the autocorrelator. One point of special interest is the role of a finite phase-matching bandwidth for the second-harmonic-generation process. According to theory, the measurement should not be critically sensitive to bandwidth restrictions or, equivalently, to group-velocity mismatch between fundamental and second-harmonic waves.[9] We have tested this point experimentally by using KDP crystals of several different thicknesses. Figure 1 shows autocorrelation traces of compressed pulses obtained with crystal thicknesses of 0.1, 0.3, and 0.5 mm. The smaller bandwidth of the 0.5-mm crystal makes the autocorrelation less sensitive to frequency components far from the center wavelength that contribute to the wings of the pulses. The 0.1- and 0.3-mm crystals, however, yield essentially identical pulse measurements. Thus we may conclude that autocorrelation measurements using a 0.1-mm KDP crystal provide an accurate pulse-width determination even for pulses as short as 16 fsec.

As an application of our compressed pulses, we have performed time-resolved reflectometry measurements of multiple-layer dielectric mirror coatings. Pulse shaping and distortion from a dielectric mirror are expected to depend not only on the reflectivity spectrum but also on the phase spectrum of the mirror. The mirror's phase spectrum is determined by the details of the dielectric-coating design and fabrication and is generally unspecified for commercial mirrors. Although in principle the phase response of a mirror coating can be measured interferometrically, time-domain measurements provide a simple, direct assessment of dispersive pulse-shaping effects.

A variety of mirror types exist that are suitable for use even with pulses as short as 16 fsec. Neither aluminum reflectors nor single-stack dielectric mirrors used near their center wavelength produced a noticeable effect on the reflected pulse shape. For example, even after 30 normal-incidence reflections from a Newport Research

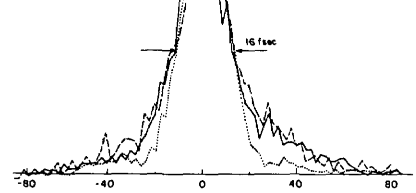

Fig. 1. Autocorrelation traces of compressed pulses obtained by background-free second-harmonic generation in KDP crystals of 0.1-, 0.3-, and 0.5-mm lengths.

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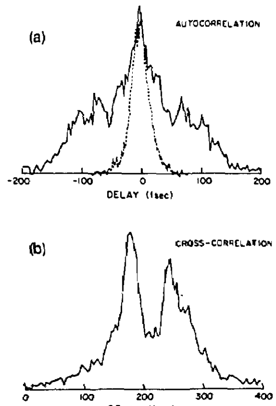

Fig. 2. (a) Autocorrelation traces of compressed pulses before and after a single, 45° incidence reflection from a typical broadband dielectric mirror. (b) Cross-correlation of reflected pulse with incident pulse.

Corporation Type ER.2 enhanced aluminum coating, no pulse distortion was evident. Broadband dielectric reflectors, however, can produce a variety of pulse-shaping effects. The exact pulse-shaping behavior depends on the details of the coating and the angle of incidence as well as on the duration, wavelength, and spectral profile of the test pulses. In many instances we have observed markedly different effects even for mirrors of nominally the same manufacture and design. In what follows we confine our attention to the case of broadband dielectric reflectors. The high reflectivity range of these mirrors extends throughout the visible; consequently, the pulse-distortion effects can be attributed exclusively to the phase response.

Figure 2(a) shows a typical measurement of pulse distortion upon reflection from a broadband dielectric mirror. The autocorrelation traces were obtained before and after a single reflection at 45° incidence from a Newport Research Corporation Type BD.1 mirror coating. The severe distortion, evidenced by the multiply peaked autocorrelation, suggests that not only quadratic but also higher-order variations of phase with frequency must be considered. This fact was confirmed by varying the separation of the grating-pair compressor; successful compensation of the distortion could not be achieved. To determine the reflected pulse shape further, we measured the cross correlation of the reflected pulse with the incident pulse by placing the test mirror in one arm of the autocorrelator. The cross-correlation trace, shown in Fig. 2(b), indicates that the reflected pulse consists of two main peaks separated by ~70 fsec and superimposed upon a broad background 150–200 fsec in duration.

Since pulse-shaping effects from broadband dielectric mirrors are strongly and directly manifest with the compressed pulses, this technique may be applied to characterize mirror coatings for femtosecond laser cavities. To demonstrate this point, we performed reflectometry measurements on two different broadband reflectors, which were also tested within the CPM dye laser. These 10-cm radius-of-curvature mirrors were each used in the CPM laser to focus both the 514.5-nm pump light and the 625-nm mode-locked pulses into the Rhodamine 6G gain medium. Both mirrors were obtained from the CVI Laser Corporation. One mirror, which we designate mirror A, had a standard EBLM10 broadband coating. The other, mirror B, had a custom-fabricated coating consisting of a cascade of two quarter-wave stacks, the outer stack centered at 619 nm and the inner at 514.5 nm. The use of mirror A in the cavity resulted in long, ~280-fsec pulses accompanied by energy fluctuations on a microsecond time scale. This behavior is commensurate with our reflectometry data. Autocorrelation traces plotted in Fig. 3, measured, respectively, before and after two near-normal-incidence reflections, plainly illustrate severe pulse broadening and demonstrate that at our wavelength mirror A is unsuitable for application in a femtosecond laser. In contrast, with mirror B the laser produced stable, short pulses of 80–85-fsec duration. For this mirror no pulse distortion is noticeable in the reflectometry data. Thus the performance of the broadband dielectric mirrors in the CPM laser may be directly related to the pulse-distortion behavior of the mirrors as measured by compressed pulses.

A qualitative explanation of observed pulse-shaping effects may be obtained by calculating the response of a typical broadband dielectric mirror design. Broadband mirrors covering the entire visible range are usually produced by cascading two highly reflecting quarter-wave stacks, one for the blue-green and one for the red wavelengths.[10] We consider a prototypical double-stack coating for normal incidence, with one quarter-wave stack centered at 480 nm and the other at 625 nm. Each stack is composed of seven pairs of alternating high- and low-index layers and one extra high-index layer. The high- and low-index materials are ZnS ($n = 2.3$) and MgF_2 ($n = 1.38$), respectively, and the substrate is BK-7 ($n = 1.52$). A low-index spacer layer, quarter-wave at the mean of the two center wavelengths (570 nm), is included between the stacks to eliminate the reflectivity dip that would otherwise be present at this wavelength.[11] With these specifications, two distinct broadband coatings are possible since the blue and red stacks can be deposited in either order.

The phase spectra for these two mirror configurations are shown in Fig. 4. In each case the reflectivity is greater than 99.9% thoughout most of the visible. For the interpretation of these phase spectra, it is important to recall that a linear variation of phase with frequency produces only a uniform pulse delay. Quadratic vari-

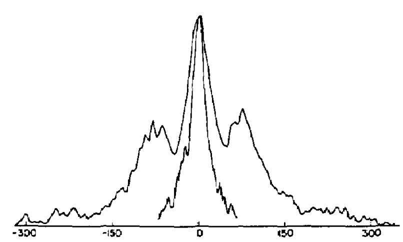

Fig. 3. Autocorrelation traces of compressed pulses before and after two normal-incidence reflections from CPM mirror A. Mirror B showed no noticeable pulse broadening.

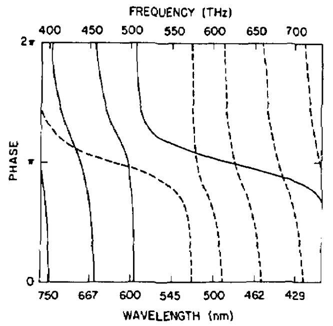

Fig. 4. Calculated phase spectra for two broadband dielectric mirror designs. Solid lines correspond to mirror with blue outer stack; dashed lines to mirror with red outer stack.

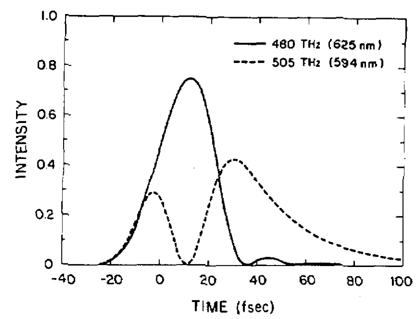

Fig. 5. Calculated reflected pulse-intensity profiles for 20-fsec FWHM Gaussian pulses incident upon broadband dielectric mirror with external blue stack shown in Fig. 4. Solid and dashed lines correspond to pulses with center frequencies of 480 and 505 THz, respectively.

ation leads to conventional dispersive broadening. Cubic and higher-order phase variations can result in a complex reshaping of the pulse. Thus, for pulses at our wavelength of 625 nm, significant distortion may occur if the highly reflecting red stack is placed close to the substrate with the blue stack on top. The blue stack introduces rapid phase variations as a function of frequency because sidelobes in its reflectivity spectrum lead to partial reflections in this outer, off-resonant stack. In the complementary configuration, the pulses are reflected from the outer red stack and do not propagate through the blue stack. In this instance the phase variation is almost linear within the pulse bandwidth, and no pulse distortion occurs.

To elucidate the reflectometry behavior further, we have calculated the pulse shapes that should result after reflection from our prototypical double-stack mirror. Figure 5 shows two representative intensity profiles calculated for 20-fsec FWHM Gaussian pulses with center frequencies of 480 and 505 THz (625 and 594 nm) after a single normal-incidence reflection from the broadband mirror with the blue stack outside. The doubly peaked profile calculated for 505 THz is similar qualitatively to many of our experimental results (see Figs. 2 and 3). Because of the highly nonlinear phase variation, the calculated reflectometry behavior is strongly dependent on the pulse center frequency. Negligible pulse distortion is produced by the complementary mirror configuration with the red stack outside. Thus the order in which the two highly reflecting dielectric stacks are deposited is found to be the most important factor in determining the pulse-shaping behavior.

In summary, we have utilized compressed pulses as short as 16 fsec to perform time-resolved reflectometry measurements of dielectric mirrors. Autocorrelation and cross-correlation techniques were used to characterize changes in pulse shape and duration. Pronounced pulse distortion, which would not be evident if longer pulses were used, was observed for several broadband commercial mirrors. We have applied our technique to broadband mirrors used in a femtosecond CPM dye laser and have found a direct relationship between the reflectometry data and the laser pulse width and stability. Finally, we have established that our experimental results are commensurate with theoretical predictions for a prototypical double-stack broadband coating.

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* Present address, Bell Communications Research, Holmdel, New Jersey 07733

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