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Title	Femtosecond-pulse laser chirp compensated by cavity-mirror dispersion
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Citation	Optics Letters, 11(8), 504-506 https://doi.org/10.1364/OL.11.000504
Issue Date	1986-08-01
Doc URL	https://hdl.handle.net/2115/45320
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Type	journal article
File Information	OL11-8_504-506.pdf

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Femtosecond-pulse laser chirp compensated by cavity-mirror dispersion

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Received March 18, 1986; accepted May 28, 1986

The duration of pulses generated from a simple colliding-pulse mode-locked cw dye laser is measured as a function of cavity-mirror dispersion. The optimum amount of mirror dispersion of $\ddot{\phi}(\omega) \cong +1.8 \times 10^{-28}$ sec^2 and a suitable mirror coating for upchirp compensation are identified. The adjustment of mirror dispersion only, without additional dispersive elements, generates continuous trains of pulses as short as 50 fsec.

Since the development of continuous trains of pulses shorter than 100 fsec from a colliding-pulse mode-locked cw (CPM) dye laser,[1] many efforts to generate shorter pulses directly from the laser have been carried out.[2] Those studies have shown that for the generation of shorter pulses, the most important thing is to compensate for the chirp arising from dispersion $\phi(\omega)$ and the phase modulation $\phi(t)$, which are due to intracavity optical elements. There are several sources of dispersion and phase modulation in the CPM laser.[3-6] Sources of dispersion include the following: (1) the dispersion that is due to multilayer dielectric mirrors, which can have positive or negative dispersion, (2) the positive group-velocity dispersion arising from the unsaturated gain of an amplifier (Rhodamine 6G; R6G) and from the use of ethylene glycol (EG) solvents and air and prism glasses, (3) the negative group-velocity dispersion arising from the unsaturated loss in an absorber [diethyloxadicarbocyanine iodide (DODCI) and its photoisomer], and (4) the negative group-velocity dispersion that necessarily accompanies the angular dispersion introduced by the prisms. Sources of phase modulation include the positive self-phase modulation arising from the transient saturation of the gain of the amplifier and from the positive nonlinear refractive indices of EG, R6G, and DODCI as well as the negative self-phase modulation arising from the transient saturation of the DODCI absorption. Dietel *et al.*[4] produced pulses shorter than 60 fsec by the adjustment of the optical path of a positive-dispersion prism glass in the cavity for compensation of chirp from negative self-phase modulation. Valdmanis *et al.*[7] recently produced pulses as short as 27 fsec by adjusting the distance between prisms, resulting in negative cavity dispersion, and compensating for chirp from positive self-phase modulation. Consequently, Dietel's[4] conclusion that downchirp is dominant in the CPM laser is contrary to Valdmanis's conclusion[7] that upchirp is dominant. In addition, the cavity configurations for those experiments complicated the optical alignment of many elements and made it difficult to determine the inherent dispersion of the CPM laser because of the additional insertion of one or four prisms, which led to negative cavity dispersion.

It is known that the pulse duration of the CPM laser depends critically on the selection of the cavity mirrors, even if they have similar high quality and high reflectivity.[8] However, the criteria for the best mirror coating for the generation of the short pulses have not been studied experimentally. Recently Silvestri *et al.*[3] evaluated the dispersion of mirrors by using a calculation of wavelength-dependent phase shifts due to dielectric multilayer mirrors for use in CPM lasers. Shortly after that, Dietel *et al.*[6] confirmed experimentally that the dispersion that is due to the mirrors is equivalent to that from the inserted prism glass with negative cavity dispersion. In this Letter we experimentally clarify the sign and the optimum amount of mirror dispersion for the compensation of chirp from the dispersion and self-phase modulation proper to the CPM laser. We also report on the production of continuous trains of pulses as short as 50 fsec in a simple cavity configuration, by the adjustment of mirror dispersion only, without additional intracavity elements.

The cavity of our CPM laser consists of a R6G jet (3.6×10^{-3} m, 233-μm thickness) to provide gain, a DODCI jet (6.8×10^{-3} m, 39-μm thickness) as an absorber, and seven multilayer dielectric mirrors, as shown in Fig. 1. The total length of the cavity is 325 cm (10.8-nsec periods). The curvatures of the pair of

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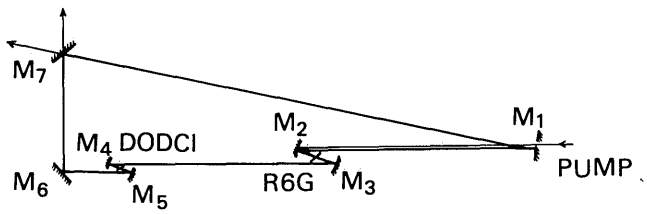

Fig. 1. Cavity configuration of the CPM laser used in the experiment. The focusing mirrors around the amplifying and absorbing jets are, respectively, M_2 and M_3 (R_c = 10 cm) and M_4 and M_5 (R_c = 2.5 cm). The perimeter of the cavity is 325 cm.

concave mirrors M_2 and M_3 around the amplifier and M_4 and M_5 around the absorber are 10 and 2.5 cm, respectively. There is tight focusing at the absorber jet to produce deep saturation. Two of three flat mirrors, M_1 (3.5° incident angle) and M_6 (45° incident angle), are changed to determine the optimum dispersion, as described below. The transmission of the output coupling mirror (M_7 or M_6) is 1 to 5% at a wavelength around 630 nm. Pump powers provided by a cw Ar-ion laser (Spectra-Physics Model 171-18) are typically 3 to 6 W at 514.5 nm. The tip of the nozzle for both dye jets is constructed from four optically polished ruby blocks to keep the jet streams uniform and stable.[9] A home-made dye circulator has a series of four accumulators, a 1-μm filter, and a high-pressure (13 kg/cm^2) magnetic gear pump. The shortest pulse duration is always obtained when the dye-laser beam is focused at the relatively thin-edge side of the absorber-jet stream. The pulse durations are measured by a usual background-free second-harmonic-generation autocorrelator (0.5-mm ADP or 0.2-mm KDP crystals)[10] operated in a fast-scan mode using a shaker and in a slow-scan mode using a motor. It is assumed that the instantaneous time variation of the pulses follows a sech^2 dependence. The pulse spectrum around 630–640 nm is monitored by an optical multichannel analyzer with a polychromator.

In general, the effect of the multilayer dielectric mirror on the amplitude and phase of an incident EM wave $E_i(\omega)$ depends on the angular frequency ω of the wave and hence on its wavelength. The reflected complex amplitude of the field $E_r(\omega)$ is described by $E_r(\omega) = r(\omega)\exp[i\phi(\omega)]E_i(\omega)$, where $R(\omega) = |r(\omega)|^2$ is the intensity reflectance and $\phi(\omega)$ is the phase shift. The reflectance and the phase shift are numerically calculated by using a matrix formulation for multilayer filters[11] with the aid of a computer. The effective quantity for chirp compensation is the second derivative of the phase shift $\ddot{\phi}(\omega) = \partial^2\phi(\omega)/\partial\omega^2$.[3] The quantity $\ddot{\phi}(\omega)$ is related to the group-velocity dispersion $\partial^2 k(\omega)/\partial\omega^2$ of a dispersive material with an effective length l by the equation $\ddot{\phi}(\omega) = -l\partial^2 k(\omega)/\partial\omega^2$. Therefore, its sign is opposite that of the group-velocity dispersion. For all the mirrors, the values of $R(\omega)$ and $\ddot{\phi}(\omega)$ in the vicinity of the lasing wavelength were calculated as a function of the wavelength for the incident angle and the p component of polarization, according to the experimental condition. In the calculation, the absorption and dispersion of the layer materials of TiO_2 (n_H = 2.25) and SiO_2 (n_L = 1.46) were neglected. All the mirrors were carefully made from uniform multilayers with a thickness variation smaller than 6%.

In order to keep the dispersion small, each of mirrors M_3, M_4, and M_5 was constructed from a single stack of 23 $\lambda_0/4$ layers (with a resonance wavelength of λ_0 = 625 nm for normal incidence). The calculated value of $\ddot{\phi}(\omega)$ near the lasing wavelength for an incident angle of order of a few degrees is about 5×10^{-31} sec^2 near the center of the reflection band and is therefore negligibly small. The M_2 mirror, for the nearly normal reflection of the dye and pump beams, was constructed from a stack doubly coated with 23 $\lambda_{0up}/4$ layers (upper air side, λ_{0up} = 625 nm) and 22 $\lambda_{0low}/4$ layers (lower substrate side, λ_{0low} = 500 nm). Similarly, the value of $\ddot{\phi}(\omega)$ is also about 5×10^{-31} sec^2 and is negligibly small. For doubly coated stack mirrors, the variation of $\ddot{\phi}(\omega)$ is influenced mainly by that of the upper stack of multilayer coatings, while the variation of $R(\omega)$ is influenced by that of both coatings. Therefore, a mirror with a specified $\ddot{\phi}(\omega)$ and $R(\omega)$ at a given wavelength can be designed by the selection of a suitable combination of the resonance wavelength of each stack of a multistack mirror. Similarly, a double-coating stack of a 23 $\lambda_{0up}/4$-layer (λ_{0up} = 630 nm) and 24 $\lambda_{0low}/4$-layer (λ_{0low} = 520 nm) mirror for normal

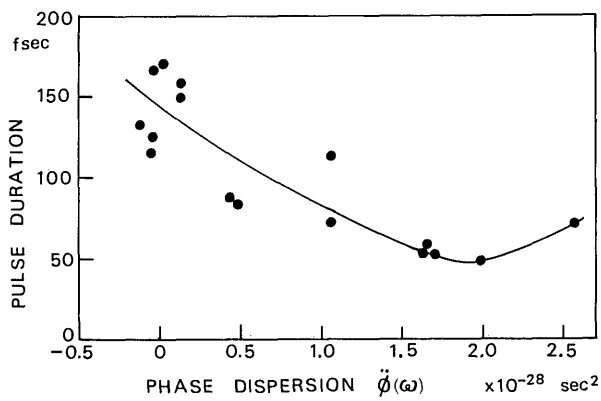

Fig. 2. Generated pulse duration as a function of cavity-mirror dispersion $\ddot{\phi}(\omega)$.

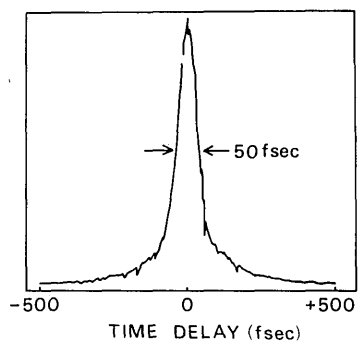

Fig. 3. Autocorrelation function of pulses from the CPM laser with chirp compensation by a cavity mirror. The full width at half-maximum corresponds to a sech^2 pulse duration of 50 fsec.

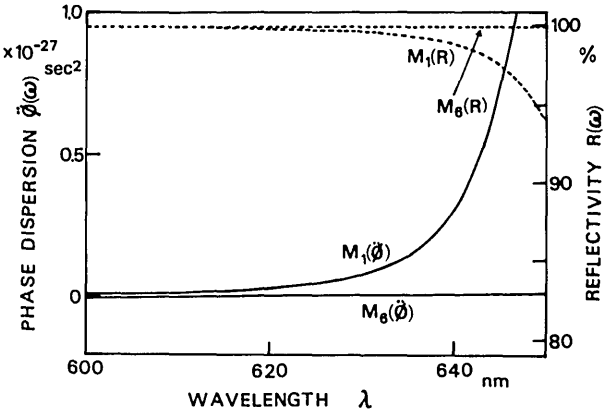

Fig. 4. The second derivative $\ddot{\phi}(\omega)$ of the phase shift and the reflectivity $R(\omega)$ plotted against the wavelength for mirrors M_1 and M_6 for the configuration used in the generation of the shortest pulse.

incidence was used for M_7. The value of $\ddot{\phi}(\omega)$ at an incident angle of 41.5° to M_7 is about 4×10^{-30} sec^2 and is also negligibly small. The transmission is about 1% near the lasing wavelength.

Intracavity dispersion was changed over a wide range from positive to negative values by using various flat mirrors for M_1 and M_6. The values of $\ddot{\phi}(\omega)$ and $R(\omega)$ as a function of the wavelength for 10 mirrors of different coatings were calculated. At each mirror configuration, the generated pulse duration was measured by carefully adjusting an operating condition, such as the pump power and the cavity alignment, while monitoring the fast scanned autocorrelation traces on an oscilloscope. Consequently, the dependence of the pulse duration on the total dispersion $\ddot{\phi}(\omega)$ of the cavity mirrors was obtained as shown in Fig. 2. The shortest pulse duration, 50 fsec (Fig. 3), with average output powers of 28 mW at the center wavelength of 636 nm (with the spectral width $\Delta\lambda$ = 7.8 nm of the nearly transform-limited value), was generated around $\ddot{\phi}(\omega) = 1.8 \times 10^{-28}$ sec^2 and not around $\ddot{\phi}(\omega) = 0$. This result implies that in a simple CPM laser, upchirp is dominant and is compensated for by negative group-velocity dispersion [corresponding to the amount of $\ddot{\phi}(\omega) = 1.8 \times 10^{-28}$ sec^2] from the cavity mirrors only. The dominant upchirp is caused by positive self-phase modulation, which is probably due to fast-response-time-dependent nonlinear refractive indices from the electronic hyperpolarizability of EG, R6G, and DODCI.[7,12] This effect occurs more remarkably in the simple CPM laser because the cavity configuration is free from negative cavity dispersion from prisms and the pulse beam to the absorber jet is tightly focused (1.25-cm focusing length).

Figure 4 shows calculated dispersion $\ddot{\phi}(\omega)$ and reflectance $R(\omega)$ curves of mirrors M_1 for 3.5° incident angle and M_6 for 45° incident angle, which were used in the generation of the shortest pulses. Mirror M_1 was made from a doubly coated stack of 13 $\lambda_{0up}/4$ layers (λ_{0up} = 535 nm) and 10 $\lambda_{0low}/4$ layers (λ_{0low} = 650 nm) for normal incidence. Mirror M_6 was made from a single stack of 25 $\lambda_0/4$ layers (λ_0 = 625 nm) for a 45° incident angle. Near the lasing wavelength both mirrors M_1 and M_6 have a similar high reflectance but much different dispersions of $\ddot{\phi}(\omega) = 1.8 \times 10^{-28}$ and 1.0×10^{-30} sec^2, respectively. We conclude that upchirp is compensated for by the negative group-velocity dispersion from mirror M_1. It should be noted that, in contrast to the expectation that a single-coating stack should be better, we find that the best mirror is the double-coating stack with a resonance wavelength of the upper-side stack considerably shorter than the lasing wavelength. The resonance wavelength of the lower-side stack should be near the lasing wavelength. We believe that because the dispersion of the mirror is rapidly increasing with wavelength (around the lasing wavelength), not only the term of the second derivative $\ddot{\phi}(\omega)$ of the phase shift but also the term of the third derivative $\dddot{\phi}(\omega)$ contributes to compensation for linear and nonlinear chirp. As was already pointed out,[5] this effect is also important for pulse shortening of the femtosecond pulse laser.

It was found that large broadening and instability of the pulses occurred when a mirror with a dispersion curve that exhibits large oscillatory variations with frequency (near the lasing wavelength) was used as one of the cavity mirrors.

In summary, we have experimentally measured the dependence of the pulse duration on cavity-mirror dispersion in a simple CPM laser. The optimum amount of dispersion and the suitable mirror coatings for compensation of upchirp were investigated. Continuous trains of pulses as short as 50 fsec were generated at a wavelength of 636 nm.

We are grateful to M. Nomura for his help with the computer calculation and A. Bodek for his critical reading of the manuscript.

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