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Femtosecond-pulse laser chirp compensated by cavity-mirror dispersion

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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 \( \phi(c) = +1.8 \times 10^{-28} \text{ sec}^2 \) 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.\(^2\) These 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 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.\(^4\) 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 \times 10^{-3} \text{ m}, 233-\text{um} thickness) to provide gain, a DODCI jet (6.8 \times 10^{-3} \text{ m}, 39-\text{um} 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
called Mirrors M2 and M3 around the amplifier and M4 and M5 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 concave mirrors M2 and M3 around the amplifier and M4 and M5 around the absorber are 10 and 2.5 cm, respectively. The focusing mirrors around the amplifying and absorbing jets are, respectively, M2 and M3 (Rc = 10 cm) and M4 and M5 (Rc = 2.5 cm). The perimeter of the cavity is 325 cm.

In order to keep the dispersion small, each of mirrors M3, M4, and M5 was constructed from a single stack of 23 λo/4 layers (with a resonance wavelength of λo = 625 nm for normal incidence). The calculated value of d2φ(ω) 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 M2 mirror, for the nearly normal reflection of the dye and pump beams, was constructed from a stack doubly coated with 23 λo/4 layers (upper air side, λo/4 = 625 nm) and 22 λo/4 layers (lower substrate side, λo/4 = 500 nm). Similarly, the value of d2φ(ω) is also about 5 × 10^{-31} sec^2 and is negligibly small. For doubly coated stack mirrors, the variation of d2φ(ω) is influenced mainly by that of the upper stack of multilayer coatings, while the variation of φ(ω) is influenced by that of both coatings. Therefore, a mirror with a specified φ(ω) and R(ω) at a given wavelength can be designed by the selection of a suitable combination of the resonance wavelength of each stack of multistack mirror. Similarly, a double-coating stack of a 23 λo/4/layer (λo/4 = 630 nm) and 24 λo/4/layer (λo/4 = 520 nm) mirror for normal

Fig. 1. Cavity configuration of the CPM laser used in the experiment. The focusing mirrors around the amplifying and absorbing jets are, respectively, M2 and M3 (Rc = 10 cm) and M4 and M5 (Rc = 2.5 cm). The perimeter of the cavity is 325 cm.

In general, the effect of the multilayer dielectric mirror on the amplitude and phase of an incident EM wave E0(ω) depends on the angular frequency ω of the wave and hence on its wavelength. The reflected complex amplitude of the field Er(ω) is described by

\[ E_r(ω) = \overline{R(ω)} \exp[0](ω) E_0(ω) \]

where \( R(ω) = \left | \overline{r(ω)} \right |^2 \) is the intensity reflectance and \( φ(ω) \) is the phase shift. The reflectance and the phase shift are numerically calculated by using a matrix formulation for multilayer filters with the aid of a computer. The effective quantity for chirp compensation is the second derivative of the phase shift \( φ(ω) = \overline{d^2φ(ω)} \overline{dω^2} \). The quantity φ(ω) is related to the group-velocity dispersion \( \overline{d^2k(ω)} \overline{dω^2} \) of a dispersive material with an effective length l by the equation \( φ(ω) = -l \overline{d^2k(ω)} \overline{dω^2} \). Therefore, its sign is opposite that of the group-velocity dispersion. For all the mirrors, the values of R(ω) and φ(ω) 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 TiO2 (nH = 2.25) and SiO2 (nI = 1.46) were neglected. All the mirrors were carefully made from uniform multilayers with a thickness variation smaller than 6%.

Fig. 2. Generated pulse duration as a function of cavity-mirror dispersion φ(ω).

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 60 fsec.
incident angle. Near the lasing wavelength both mirrors \( M_1 \) and \( M_6 \) have a similar high reflectance but much different dispersions of \( \varphi(\omega) = 1.8 \times 10^{-28} \) and \( 1.0 \times 10^{-30} \text{ sec}^{-2} \), 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 \( \varphi(\omega) \) of the phase shift but also the term of the third derivative \( \varphi(\omega) \) contributes to compensation for linear and nonlinear chirp. As was already pointed out,\(^1\) 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.

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References