Femtosecond-pulse laser chirp compensated by cavity-mirror dispersion

Mikio Yamashita
Laser Research Section, Radio- & Opto-Electronics Division, Electrotechnical Laboratory, Tsukuba, Ibaraki, Japan

Mitsuru Ishikawa
Tsukuba Laboratory, Hamamatsu Photonics, Tsukuba, Ibaraki, Japan

Kenji Torizuka and Takuzo Sato
Laser Research Section, Radio- & Opto-Electronics Division, Electrotechnical Laboratory, Tsukuba, Ibaraki, Japan

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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 $\phi(c) = +1.8 \times 10^{-28}$ 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, many efforts to generate shorter pulses directly from the laser have been carried out. 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. 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 [diethyloxadicyanide 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. 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. Consequently, Dietel's conclusion that downchirp is dominant in the CPM laser is contrary to Valdmanis's conclusion 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. However, the criteria for the best mirror coating for the generation of the short pulses have not been studied experimentally. Recently Silvestri et al. 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. 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 ($3.6 \times 10^{-3}$ m, 233-µm thickness) to provide gain, a DODCI jet ($6.8 \times 10^{-3}$ 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
Concave mirrors M2 and M3 around the amplifier and M4 and M6 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, M1 (3.5° incident angle) and M6 (45° incident angle), are changed to determine the optimum dispersion, as described below. The transmission of the mirrors, M1 (3.5° incident angle) and M6 (45° incident angle), are changed to determine the optimum dispersion, as described below. The transmission of the output coupling mirror (M7 or M6) 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. A home-made dye circulator has a series of four accumulators, a 1-μm filter, and a high-pressure (13 kg/cm²) 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) 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 pulse follows a sech² 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 \( E_\omega \) depends on the angular frequency \( \omega \) of the wave and hence on its wavelength. The reflected complex amplitude of the field \( E_r(\omega) \) is described by \( E_r(\omega) = r(\omega)exp[i\phi(\omega)]E_\omega(\omega) \), 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 with the aid of a computer. The effective quantity for chirp compensation is the second derivative of the phase shift \( \dot{\phi}(\omega) = \partial^2 \phi(\omega)/\partial \omega^2 \). The quantity \( \phi(\omega) \) 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 \( \dot{\phi}(\omega) = -iL^2\ddot{k}(\omega)/\partial \omega^2 \). Therefore, its sign is opposite that of the group-velocity dispersion. For all the mirrors, the values of \( R(\omega) \) and \( \dot{\phi}(\omega) \) 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 M3, M4, and M5 was constructed from a single stack of 23 \( \lambda_0/4 \) layers (with a resonance wavelength of \( \lambda_0 = 625 \) nm for normal incidence). The calculated value of \( \dot{\phi}(\omega) \) near the lasing wavelength for an incident angle of order of a few degrees is about \( 5 \times 10^{-31} \) sec² near the center of the reflection band and is therefore negligibly small. The M3 mirror, for the nearly normal reflection of the dye and pump beams, was constructed from a stack doubly coated with 23 \( \lambda_0/4 \) layers (upper air side, \( \lambda_{up} = 625 \) nm) and 22 \( \lambda_{low}/4 \) layers (lower substrate side, \( \lambda_{low} = 500 \) nm). Similarly, the value of \( \dot{\phi}(\omega) \) is also about \( 5 \times 10^{-31} \) sec² and is negligibly small. For doubly coated stack mirrors, the variation of \( \dot{\phi}(\omega) \) 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 \( \dot{\phi}(\omega) \) and \( R(\omega) \) at a given wavelength can be designed by the selection of a suitable combination of the resonance wavelengths of each stack of a multistack mirror. Similarly, a double-coating stack of a 23 \( \lambda_{up}/4 \)-layer (\( \lambda_{up} = 630 \) nm) and 24 \( \lambda_{low}/4 \)-layer (\( \lambda_{low} = 520 \) nm) mirror for normal
incident angle. Near the lasing wavelength both mir-

rors $M_1$ and $M_6$ have a similar high reflectance but
much different dispersions of $\phi(\omega) = 1.8 \times 10^{-28}$ and
$1.0 \times 10^{-30}$ sec$^2$, respectively. We conclude that up-
chirp 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 double, 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 deriva-
tive $\phi(\omega)$ of the phase shift but also the term of the
third derivative $\phi(\omega)$ contributes to compensation for
linear and nonlinear chirp. As was already pointed
out, 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. Con-

tinuous trains of pulses as short as 50 fsec were gener-
ated at a wavelength of 636 nm.

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