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A Chirp-Compensation Technique Using Incident-Angle Changes of Cavity Mirrors in a Femtosecond Pulse Laser

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Abstract—A technique for chirp-compensation in a CPM laser is presented. By using the change of the incident angle to multilayer dielectric cavity mirrors, the intracavity second-order dispersion $\ddot{\phi}(\omega)$ is adjusted without any additional elements. It is confirmed that the optimum value of $\ddot{\phi}(\omega) = +2.1 \times 10^{-28} \text{ s}^2$ obtained when up-chirp was compensated and pulses as short as 55 fs were generated is reasonable, by comparison to analytic results of chirp behaviors. In addition, the effect of the third-order dispersion $\dddot{\phi}(\omega)$ at the optimum value of $\ddot{\phi}(\omega)$ on pulses is evaluated.

RECENT studies on a colliding-pulse mode-locked (CPM) CW dye laser showed that the most important thing for femtosecond pulse generation is to compensate for the frequency chirp arising from dispersion and self-phase modulation in the cavity. For the chirp compensation, there are presently four techniques as follows: 1) the down-chirp compensation by the adjustment of the optical path in a prism inserted in the cavity [1], 2) the up-chirp compensation by the adjustment of the distance between four prisms inserted in the cavity [2], 3) the up-chirp compensation by the adjustment of the incident angle to a pair of Gires-Tournois interferometers used as parts of cavity mirrors [3], and 4) the up-chirp compensation by the use of the optimum second-order dispersion (the details will be described below) due to $\lambda_0/4$ multilayer dielectric mirrors composing of the cavity [4]. The former three techniques necessitate the additional optical elements which furthermore complicate the optical alignment of the CPM ring laser. On the other hand, the last one has the inconvenience that cavity mirrors must be exchanged to other mirrors of different coatings to determine the optimum dispersion. In this paper we report that, by the change of the incident angle to cavity mirrors instead of the exchange of mirrors in the last technique, up-chirp can be compensated. In addition, it is shown that the amount of the second-order dispersion obtained for the up-chirp compensation agrees well with a recent analytic result of chirp behaviors due to intracavity self-phase modulation [5]. Furthermore, the influence of the third-order disper-

sion due to mirrors at the optimum amount of the second-order dispersion on femtosecond pulses is estimated.

The simple CPM (R6G + DODCI) laser used without additional elements is nearly identical to the one we previously described [4], except for the following points. The intracavity second-order dispersion was varied by moving mirrors M_6 and M_7 to the opposite direction of each other, as shown by arrow bars in Fig. 1(a). That is, the incident angles to mirrors M_6 and M_7 , respectively, were varied from 45 to 60° and from 40 to 25°. The details of the mirror coatings will be described below. The transmission of the output coupling mirrors (M_7 or M_1) is about 0.5 ~ 1.0 percent around 630 nm. Pump powers of 2.4–3.2 W at 514.5 nm were provided by a CW Ar ion laser. The DODCI concentration was $5 \times 10^{-3} \text{ M/l}$ in a jet from a 39 μm wide nozzle slit. Typical average output power was 10 mW. The femtosecond pulse durations were measured by a background-free SHG autocorrelator (0.2 mm KDP crystal) operated in a fast-scan mode and a slow-scan mode. Hyperbolic secant-squared pulse shapes were assumed throughout in driving the pulse durations from the full width at half maximum of the intensity autocorrelation functions. The pulse spectra around 625–640 nm were monitored on an oscilloscope using an optical multichannel analyzer.

The phase of the EM wave of the laser pulse is shifted when the wave is reflected by a $\lambda_0/4$ multilayer dielectric mirror. The phase shift $\phi(\omega)$ is a function of the optical angular frequency ω and can be calculated using a matrix formulation [6]. The effective phase-dispersion for chirp compensation is the second-order angular-frequency derivative $\ddot{\phi}(\omega)$ of the phase shift (the second-order dispersion). The quantity $\ddot{\phi}(\omega)$ is related to the group-velocity dispersion $\ddot{\kappa}(\omega)$ of a dispersive material with a length l by an equation $\ddot{\phi}(\omega) = -l\ddot{\kappa}(\omega)$ ($\kappa(\omega)$: wavenumber). Therefore, the positive second-order dispersion $\ddot{\phi}(\omega)$ corresponds to the negative group-velocity dispersion $\ddot{\kappa}(\omega)$. The value of $\ddot{\phi}(\omega)$ for the reflected laser pulse with a given angular frequency depends on the incident angle θ to a mirror and the resonance wavelength λ_0 of the multilayer stack of its mirror.

When the incident angle θ is larger than 45° and the wavelength λ_d of the incident laser-pulse is relatively longer than λ_0 , the $\ddot{\phi}(\omega)$ has a large value (10^{-29} – 10^{-27}

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Fig. 1. (a) Cavity configuration of the simple CPM laser used. Two arrow bars represent the direction of the movement of mirrors M_6 and M_7 for the change of the incident angle θ of laser beams. (b) The second-order dispersion $\ddot{\phi}(\omega)$ and the reflectivity $R(\omega)$ at 635 nm plotted against the incident angle θ to mirrors M_6 (45 to 60°) and M_7 (40 to 25°).

s^2) in the positive region and increases with the increase of the angle θ . Our previous study showed that in a simple CPM laser up-chirp is dominant and compensated by a large value of $\ddot{\phi}(\omega) \approx +1.8 \times 10^{-28} s^2$ in the positive region. Therefore, mirror M_6 where the angle θ is varied from 45 to 60°, should be made from two quarter-wave stacks to produce an increasingly positive value of $\ddot{\phi}(\omega)$ (around $10^{-28} s^2$) with keeping high reflectivity $R(\omega)$. That is, the resonance wavelength $\lambda_{0,up}$ of the upper stack (the air side) for the incident angle $\theta = 45\text{--}60^\circ$ should be relatively shorter than λ_d , so as to produce a value of $\ddot{\phi}(\omega) \approx 10^{-28} s^2$ in the positive region. The resonance wavelength $\lambda_{0,low}$ of the lower stack (the substrate side) for the angle should be near λ_d , so as to keep high reflectivity. On the basis of these considerations, mirror M_6 was carefully made from two quarter-wave stack superimposed with $17 \lambda_{0,up}/4$ layers ($\lambda_{0,up} = 563$ nm) and $25 \lambda_{0,low}/4$ layers ($\lambda_{0,low} = 653$ nm) for 60° incident angle. The coated layer materials of high and low refractive indexes were TiO_2 ($n_H = 2.25$) and SiO_2 ($n_L = 1.46$). Fig. 1(b) shows calculated dependences of the second-order dispersion $\ddot{\phi}(\omega)$ and reflectivity $R(\omega)$ on the incident angle θ (between 45 and 60°) to mirror M_6 for the p -polarization beam at 635 nm.

As for mirror M_7 , a mirror causing a small value of $\ddot{\phi}(\omega)$ around λ_d is suitable, in order to keep its value almost constant at the incident angle between $\theta = 40$ and 25° . Therefore, as the incident angle dependences of $\phi(\omega)$ and $R(\omega)$ are also shown in Fig. 1(b), a mirror which had been made from two quarter-wave stacks of $23 \lambda_{0,up}/4$ -layer ($\lambda_{0,up} = 630$ nm) and $24 \lambda_{0,low}/4$ -layer ($\lambda_{0,low} = 520$ nm) for normal incidence, was used as mirror M_7 between $\theta = 40$ and 25° .

For other mirrors M_1 – M_5 , the same mirrors as the previously employed ones [4] were used. Mirrors from M_2 to M_5 cause only a negligible small value of $\ddot{\phi}(\omega) \approx 5 \times 10^{-31} s^2$, while mirror M_1 causes a constant positive value of $\ddot{\phi}(\omega) \approx +0.8 \times 10^{-28} s^2$ at about 630 nm.

The effect on the pulse of the incident angle θ to cavity mirrors was experimentally examined by the simultaneous movement of mirrors M_6 and M_7 along the directions of the angle from 45 to 60° and angle from 40 to 25°, [along arrow bars in Fig. 1(a)], respectively. At each 8 mm movement on translation stages, an optimum operating condition such as the pumping power and the alignment of the optical components was carefully adjusted, while monitoring the fast scanned autocorrelation traces on an oscilloscope. Consequently, the pulse duration dependence on the second-order dispersion $\ddot{\phi}(\omega)$ due to all cavity mirrors was obtained as shown in Fig. 2. As the value of $\ddot{\phi}(\omega)$ increases from 0.8×10^{-28} to $2.1 \times 10^{-28} s^2$, the pulse duration decreases from 90 to 55 fs, and then the further increase of $\ddot{\phi}(\omega)$ to $2.5 \times 10^{-28} s^2$ broadens the pulse duration to 68 fs. The obtained dependence and the value of $\ddot{\phi}(\omega)$ where the shortest pulses were generated, are in agreement with those in the previous experiment based on mirror exchange [4]. This indicates that the negative group-velocity dispersion corresponding to the above value of $\ddot{\phi}(\omega)$ can compensate for up-chirp mainly due to the effect of the nonlinear refractive index of the solvent of the DODCI absorber, ethylene glycol. We shall show below that the amount of this optimum dispersion is reasonable.

Miranda *et al.* carried out a simplified analysis of frequency-chirp $C(t) = \partial \delta \omega(t) / \partial t$ imposed on femtosecond pulses interacting with a saturable absorber including the effect of nonlinear refractive index due to the solvent [5]. Their results showed that the chirp in a CPM laser remarkably depends on the intracavity pulse energy E_p . In the region of $E_p \lesssim 1$ nJ, down-chirp due to the transient saturation of the absorption is dominant around the pulse peak, while in the region of $E_p \gtrsim 10$ nJ, up-chirp due to the time-dependent refractive index of the solvent is dominant. In the present CPM laser, E_p is estimated to be ~ 15 nJ, and, hence, up-chirp $C(0) > 0$ at the pulse peak occurs. By using their numerical result for $C(0)$, which was obtained as a function of the pulse duration with a parameter E_p , $C(0)$ for the present CPM laser is estimated to be $\sim 10^{+26} \text{ rad} \cdot s^{-2}$. Therefore, the amount of the second-order dispersion $\ddot{\phi}(\omega)$ necessary for this up-chirp compensation can be evaluated to be $\sim 10^{-28} s^2$ at the pulse duration of $T_{in} = 55$ fs using an equation $\ddot{\phi}(\omega) = C(0)^{-1} / [1 + (8 \times (\ln 2)^2 / (C(0) \times T_{in}^2))]^2$ [7]. This agrees, in order of magnitude, with the experimental value.

Finally, we estimate the effect of the third-order angular-frequency derivative $\dddot{\phi}(\omega)$ of the phase shift (the third-order dispersion) due to mirrors on femtosecond pulses in the present CPM laser. The value of $\dddot{\phi}(\omega)$ due to all mirrors at the above obtained optimum value of $\ddot{\phi}(\omega)$ for up-chirp compensation is calculated to be $\sim 1 \times 10^{-41} s^3$. A few research groups derived an equation describing pulse broadening due to the third-order dispersion [8], [9]. Their results showed that for the input chirp-compensated pulse, the behavior of its output pulse shape is symmetric in respect to the sign of $\ddot{\phi}(\omega)$. As the value

Fig. 2. Pulse duration as a function of the second-order dispersion $\ddot{\phi}(\omega)$ due to cavity mirrors, which was varied by the change of the incident angle to mirrors.

of $\ddot{\phi}(\omega)$ becomes large, the peak intensity of the main pulse decreases and some small pulses follow after the main pulse. From the application of their analysis to femtosecond pulses, it is found that when chirp-compensated 50 fs pulses are reflected by mirrors of $\ddot{\phi}(\omega) = 2 \times 10^{-41} \text{ s}^3$, they are hardly broadened. However, when 30 fs pulses are reflected by the same mirrors, they are broadened by about two times, and when being reflected by mirrors with less than $\ddot{\phi}(\omega) = 4 \times 10^{-42} \text{ s}^3$ they are hardly broadened. Therefore, for generation of pulses shorter than 50 fs, mirrors having $\ddot{\phi}(\omega) \leq 4 \times 10^{-42} \text{ s}^3$ should be used.

In conclusion, it has been demonstrated as a technique of chirp-compensation in a CPM laser that the change of the incident angle to cavity mirrors has enabled us to adjust the intracavity second-order dispersion and, hence, to compensate for up-chirp and generate pulses as short as 55 fs. In addition, the evaluation on the effect of the third-order dispersion due to mirrors at the optimum second-order dispersion has been done.

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