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Third-order dispersion in a passively mode-locked continuous-wave dye laser

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The pulse duration $t_p$ of a colliding-pulse mode-locked dye laser is measured versus the intracavity third-order dispersion (TOD), where the group-velocity dispersion is optimized by a four-prism sequence. The intracavity TOD is varied by the addition of layers on quarter-wave dielectric-multilayer mirrors. The shortest pulse is observed at a nonzero TOD. The TOD dependence of $t_p$ is explained by a new model that describes the saturation of absorption and gain dyes in the frequency domain, with the assumption of a steady sech$^2$-shaped pulse with weak chirp and a negligible fast self-phase modulation.

1. INTRODUCTION

Recent advances in the development of femtosecond pulse lasers showed that tuning the intracavity group-velocity dispersion (GVD) effectively shortens the pulse duration $t_p$. The tuning is done by means of two kinds of dispersive device, refractive (a glass plate or Brewster prisms) and interferometric (a Gires–Tournois (GT) interferometer or periodic dielectric-multilayer coatings on cavity mirrors). The circulating pulse in a passively mode-locked cw dye laser is formed and modified by the saturable gain and absorption, the intracavity dispersion, and an intensity-dependent refractive index of the dye solvent. The gain and absorption are the sources of the slow self-phase modulation (SPM), and the refractive index is the source of the fast SPM. The GVD-tuning devices compensate for not only GVD that is due to the intracavity elements but also a linear chirp that is due to the dominant fast SPM, as was shown by Valdmanis and co-workers.

Analysis of pulse generation in a passively mode-locked dye laser is generally quite complicated. Many theoretical studies were carried out for stationary mode-locking by pulse amplitude modulation, an effect of the intracavity GVD and the fast SPM. Furthermore, it was shown that a critical analysis of nonstationary mode locking and soliton-like pulse formation is necessary when the fast SPM is significant. When the fast SPM is weak, both saturable dyes form a stationary pulse train.

Several authors pointed out that pulse shortening is limited by the frequency-dependent GVD (the higher-order dispersion). The lowest-order term of the frequency-dependent GVD is called the third-order dispersion (TOD). Kühnike et al. first observed the effect of the frequency-dependent GVD on $t_p$ and the pulse fluctuation in the colliding-pulse mode-locked (CPM) laser that contains both the four-prism sequence and the GT interferometers. However, the frequency-dependent GVD of their GT interferometers was so strong that it was not accounted for by TOD alone. Therefore the exact TOD dependence of $t_p$ was not obtained.

Recently the numerical simulation by de Barros et al. showed that the optimization of TOD shortens the pulses of a GVD-adjusted CPM laser. The more recent experimental work by Goto et al. resulted in the generation of a 22–25-fs pulse with a combination of the cavity mirrors and the four-prism sequence. Salin et al. studied the TOD effect in soliton-like pulse formation. In spite of these investigations, a clear quantitative relation between the intracavity TOD and passive mode-locking parameters has not yet been established experimentally and theoretically.

In this paper the TOD dependence of $t_p$ is measured with good reproducibility in a GVD-adjusted CPM dye laser during weakly saturated absorption by using a combination of the cavity mirrors with additional layers for TOD tuning and the four-prism sequences. It is found that the pulse duration is broadened by a large positive and negative TOD and is minimized at nonzero TOD. The measured TOD dependence is explained by a new analysis with a frequency-domain model for a weakly chirped pulse with a sech$^2$ shape. The stationary mode locking under the weak fast SPM is treated by the model. It is shown that the TOD that generates the shortest pulse becomes of nonzero value because of the frequency-dependent phase delay caused by dye saturations and a finite band-limiting loss.

2. EXPERIMENTS

A. Tuning of Intracavity Third-Order Dispersion

First we describe the tuning of the intracavity dispersion of the CPM laser used for the experiment, where specially designed cavity mirrors and a sequence of four Brewster prisms (B.P.'s) are employed as dispersion-tuning elements (Fig. 1). The phase delay caused by these dispersive elements for one turn of the ring cavity is denoted as $\varphi(\omega)$. It is expanded to a Taylor series as $\varphi(\omega) = \varphi(0) + \varphi(1)\Delta\omega + [\varphi(2)/2]\Delta\omega^2 + [\varphi(3)/6]\Delta\omega^3 + \delta\varphi(\omega)$, where $\varphi(n)$ is the nth-order derivative of the phase delay with respect to the angular frequency $\omega$ ($n = 0, 1, 2, 3$), $\delta\varphi(\omega)$ is the higher-order term that is due to the mirrors and prism sequence, and $\Delta\omega = \omega - \omega_0$ with lasing center angular frequency $\omega_0$. Accordingly, $\varphi(0)$ and $\delta\varphi(\omega)$ are $\varphi(0) = \Sigma\varphi(n)$ and $\delta\varphi(\omega) = \delta\varphi(0) + \Sigma\varphi(n)$.
and $\delta \varphi = \delta \varphi_{ps} + \Sigma_{i} \delta \varphi_{i}$, where the subscripts $ps$ and $i$ specify the contributions of the prism sequence and each of the cavity mirrors. The dispersive contributions of the transient gain and absorption are excluded from $\varphi(\omega)$. Following the notation of Fork et al., GVD (the quadratic phase) and TOD (the cubic phase) are proportional to $-\varphi^{(2)}$ and $-\varphi^{(3)}$, respectively.

The second-order dispersion and TOD’s of the four-prism sequence ($\varphi_{ps}^{(2)}$ and $\varphi_{ps}^{(3)}$) are calculated to be $3.1 \times 10^{-3}$ rad $52 \times 10^{-3}$ fs$^2$ and $4.7 \times 10^{-3} - 24 \times 10^{-3}$ fs$^3$ at 632 nm, respectively. Here $l_p$ (mm) and $l_m$ (mm) are the prism separation and the total material length through the fused-quartz prisms. Because of the finite beam radius and spectral width of the laser, $l_m$ must be greater than a few millimeters. Therefore $\varphi_{ps}^{(2)}$ is positive ($\geq 300$ fs$^2$) when $\varphi_{ps}^{(2)} > 0$. In the experiments $l_m$ is estimated by the observation of spacings between two beams reflected from the surfaces of each prism. The variation of $l_m$ is measured more precisely by displacements of the translation stages for prism insertion.

Our dispersion-designed mirrors consist of double stacks of additional dispersive layers (upper side) and high-reflection quarter-wave layers for the lasing frequency (lower side). Each mirror is designed for use at 45° ($p$ polarization) or 0°. At 632 nm mirror number $i$ satisfies $\varphi_{i}^{(2)} > 0$, $\varphi_{i}^{(3)} < 0$, and $\delta \varphi_{i} = 0$.

In all the mirrors TiO$_2$ is used as a high-refraction material with refractive index $n_{H}$ and SiO$_2$ as a low-refraction material with refractive index $n_{L}$. The multilayer theory predicts that a double-stacked mirror with an upper stack of periodic layers, $[H(0.188\lambda_{o}) - L(0.188\lambda_{o})]^3$, and a lower stack of high-reflection layers, $[H(\lambda_{o}/4) - L(\lambda_{o}/4)]^4$, will give a dispersion comparable with that of a GT interferometer, $[H(0.25\lambda_{o}) - L(\lambda_{o})] - H(\lambda_{o}/4) - L(\lambda_{o}/4)]^4$. Here $H(n_{H}d_{H})$ and $L(n_{L}d_{L})$, respectively, denote the high- and low-refraction layers with the optical thicknesses $n_{H}d_{H}$ and $n_{L}d_{L}$, and $\lambda_{o}$ is the lasing center wavelength at ~632 nm. We use this double-stacked mirror because it can be easily manufactured by mirror suppliers with a guaranteed quality. Careful attention in the manufacturing process is also paid to the accuracy of the layer thickness and the exclusion of impurities in order to minimize dispersion error and additional absorption.

We discuss $\delta \varphi(\omega)$ of mirrors $a$, $a'$, and $b$, for the TOD tuning, whose layer compositions were described previously. Mirrors $a$, $a'$, and $b$ are composed of double stacks of

$$[L(0.206\lambda_{o}) - H(0.206\lambda_{o})]^3 - [L(0.264\lambda_{o}) - H(0.264\lambda_{o})]^{12},$$

$$[L(0.177\lambda_{o}) - H(0.177\lambda_{o})]^3 - [L(0.248\lambda_{o}) - H(0.248\lambda_{o})]^{12},$$

$$[L(0.193\lambda_{o}) - H(0.193\lambda_{o})]^3 - [L(0.237\lambda_{o}) - H(0.237\lambda_{o})]^{12},$$

respectively. In Fig. 2 the frequency dependences of $\varphi^{(2)}$ and $\varphi^{(3)}$ for mirrors $a$ and $b$ are shown. Mirrors $a$ and $b$ give the largest $|\varphi^{(2)}|$ of all the mirrors for the incident angles of 45° and 0°, respectively. The dispersion characteristics of mirror $a'$, employed for the nearly normal incidence, correspond to those of mirror $a$ at 45°. Figure 3 shows the frequency dependence of $\delta \varphi(\omega)$ at a central frequency $\omega_{o} = 2.98 \times 10^{15}$ rad/s for mirrors $a$ and $b$. The same combinations of mirrors $a$, $a'$, and $b$ for $M_1-M_4$ in Fig. 1 are employed to vary the value of the TOD, keeping $\Sigma_{i} \delta \varphi_{i}(\omega)$ minimal. It will be confirmed that $\delta \varphi(\omega)$ is negligibly small within the bandwidths of the mode-locked laser pulses, as is shown in Subsection 2.B. However, it should be noted that $\delta \varphi(\omega)$ is not sufficiently small for the shortest pulses of a GVD-adjusted CPM dye laser. For example, to obtain a 30-fs pulse, one must tune $\varphi^{(2)}$ and $\varphi^{(3)}$ within 1 fs$^2$ and 100 fs$^3$, respectively. At the same time, $\delta \varphi(\omega)$ should be less than ~6 x 10$^{-5}$ rad within the corresponding bandwidth.

B. Measurement of the Third-Order-Dispersion Dependence of the Pulse Duration

The cavity of the CPM laser that is used for the experiment (Fig. 1) consists of a pair of $f = 50$ mm concave mirrors, with a mixture of Rhodamine 6G (Rh6G) and Kiton Red as a gain medium, a pair of $f = 25$ mm concave mirrors with diethylcloxacarbocyanine iodide (DODCI) as an absorber medium, a four-prism sequence, and the five

![Fig. 1. Schematic diagram of the cavity configurations. The ring cavity-length is ~4 m (Abs., absorber).](image)

![Fig. 2. Calculated frequency dependencies of the second-order dispersion $\varphi^{(2)}$ (dashed curves) and the TOD’s $\varphi^{(3)}$ (solid curves) of TOD-tuning mirrors a and b.](image)

![Fig. 3. Higher-order dispersions in the expansions at frequency $\omega_{o} = 2.98 \times 10^{15}$ rad/s. Mirrors a and b are the same as those in Fig. 2.](image)
plane mirrors $M_1$-$M_2$. Mirrors $M_1$, $M_2$, and $M_3$ are used at nearly normal incidence. Mirrors $M_3$ and $M_4$ (the output coupler) are used at 45° ($p$ polarization). The dispersion of the all-concave mirrors is negligibly small. The output coupler contributes a constant small dispersion ($\varphi _c^{(2)} = 3.2 \text{ fs}^2$ and $\varphi _c^{(3)} = 31 \text{ fs}^3$). An angle-tuning technique is not used in this experiment. The dispersion-designed mirrors $a$, $a'$, and $b$ are employed for $M_1$-$M_4$.

The transmission $T$ of the output coupler is 3% at 632 nm, and its minimum is at 600 nm. Such a slightly blue-shifted output coupler gives stable mode locking and good reproducibility in the TOD-dependence measurement. The addition of Kiton Red to the Rh6G solution decreases the laser threshold and the circulating pulse energy. The average output is 7 mW, with 1.5 W (514.5 nm) for pumping at the 1.5-mM DODCI concentration. The jet-sheet thicknesses are 150 $\mu$m for the gain medium and 40 $\mu$m for the absorber. For this operating condition the effective dyes.}

The total material length $l_m$ of the prisms is adjusted for the GVD compensation and the generation of the shortest pulse while we monitor the spectrum and autocorrelation trace. The shortest pulse is observed slightly before the abrupt change of the spectral and correlation shapes occurs. Figure 4 shows $t_\phi$ versus $\varphi _c^{(2)}$, where $l_m$ is adjusted to generate the shortest pulse and optimize the intracavity GVD at each point of the various TOD's. Each data point indicates the average of several sets of slow-scan autocorrelation measurements. The value of $t_\phi$ is the FWHM of the intensity on the assumption of a nonchirped sech$^2$-shaped pulse. The open circles in Fig. 4 represent the TOD dependence measured at the 1.5-mM DODCI concentration by the combination of dispersive mirrors $a$, $a'$, and $b$ with prism separation $l_p = 380 \text{ mm}$. The triangles (1.0-mM DODCI concentration) and the crosses (3.80-mM DODCI concentration) represent the data measured at $l_p = 450 \text{ mm}$. For the filled circles (1.5-mM DODCI) $l_p$ and $l_m$ are varied, and $M_1$-$M_4$ are the quarter-wave mirrors with negligible dispersions. For all the data points the center wavelength is 632 $\pm$ 2 nm with a bell-shaped pulse spectrum, and the width is near the transform limit of a nonchirped sech$^2$ pulse (the measured widest spectral width is 9.1 nm).

The result shows that, in the negative-$\varphi _c^{(2)}$ region, pulse duration $t_\phi$ increases as $\varphi _c^{(2)}$ decreases (the open circles in Fig. 4). The curves (the crosses and the filled circles in Fig. 4) indicate that the minimum $t_\phi$ exists at the positive $\varphi _c^{(2)}$ (negative TOD). The optimum value, $\varphi _c^{(2)}$, is $+1000$ $\sim 1500 \text{ fs}^2$. When $\varphi _c^{(2)}$ is larger than $\varphi _c^{(2)}$, however, the tails of the autocorrelation traces of the generated pulses gradually increase as $\varphi _c^{(2)}$ increases. The reliability of the data is confirmed by the fact that all the TOD dependences of $t_\phi$ are repeatedly obtained within an accuracy of $\pm 5\%$ (the error bar) when all the dyes are fresh. Furthermore, the result is reproduced when the whole laser cavity is taken to pieces and reconstructed.

There are no measurable differences among the laser thresholds for the different combinations of the dispersive mirrors after the optimization of $\varphi _c^{(2)}$ by the prism insertion. Therefore the additional loss that is due to the dispersive layers is considered to be sufficiently small. The optimum intracavity GVD is nearly constant at any DODCI concentration in the measured $\varphi _c^{(2)}$ range. The adjustment of $l_m$ for the shortest pulse compensates for the difference of $\varphi _c^{(2)}$ between the exchanged mirrors. This fact also confirms that the practical dispersions of the used mirrors coincide with the designed values.

3. THEORETICAL CONSIDERATIONS

Generally the relation between the pulse duration ($t_\phi$) and the intracavity TOD ($-\varphi _c^{(2)}$) is treated by a numerical simulation, since a passively mode-locked laser involves a balance of many factors. However, the situation for our experiment makes it possible to analyze the experimental result by a following simplified model in the frequency domain. That is, under the present TOD-dependence measurement, pulse shaping by the fast SPM is weak. Thus the TOD-dependence of the pulse duration should be explained by the contributions of both saturable dyes.

A. Fundamental Equations in the Frequency Domain

In the group-velocity coordinate the electric field $\Psi(t)$ of the sech$^2$ pulse in the slowly varying envelope approximation is represented as

$$\Psi(t) = E(t)\exp(i\omega_0 t) + E^*(t)\exp(-i\omega_0 t),$$

with

$$E(t) = E_0[\text{sech}(t/\tau)]\exp[i\Phi(t/\tau)],$$

where $\Phi(t/\tau)$ is the slowly varying phase and $\tau = t_\phi/1.76$. In the following discussion a weak-chirp model is adopted:

$$\exp[i\Phi(t/\tau)] = 1 + \left[ c_2 \left( \frac{1}{\tau} \right)^2 + c_3 \left( \frac{1}{\tau} \right)^3 \right].$$

Here $c_2$ and $c_3$ denote the linear- and parabolic-chirp coefficients, respectively, satisfying $|c_2/2|, |c_3/6| << 1$. 

![Diagram](chart.png)
Fig. 5. Schematic diagram of the analytical model. The complex propagators of the saturable gain $\gamma_g$, the saturable absorption $\gamma_a$, and the nonlinear refractive material $\gamma_n$ are dependent on the pulse amplitude and phase. The cavity loss is mainly due to the transmission $\Sigma_i t_i$ of the mirrors. The phase delay of the mirrors $\Sigma_i \phi_i$ and the prism sequence $\phi_p$ are controlled in the experiments.

A Fourier component $\tilde{E}(\omega)$ of the field amplitude is represented as a function of the detuning $\Delta \omega (= \omega - \omega_0)$, 

$$\tilde{E}(\omega) = \int_{-\infty}^{\infty} E(t) \exp(-i \Delta \omega t) \, dt.$$ 

When the pulse passes through each optical element, $\tilde{E}(\omega)$ is modified to 

$$\tilde{E}'(\omega) = \tilde{E}(\omega) \exp[\gamma(\omega)]$$

where $\gamma(\omega) = \gamma_g, \gamma_a, \gamma_n$ are the complex propagators in the frequency domain (Fig. 5). Here $\gamma_g(\omega), \gamma_a(\omega), \gamma_n(\omega)$ represent, respectively, the effect of the saturable gain, the saturable absorption, the fast SPM, and the contribution from the other optical components, including the dispersion-tuning elements.

Assuming that the pulse is only slightly altered with regard to any one element on a single pass, we treat only the lowest-order term for the propagators. This means that strong modulation phenomena, such as solitonlike pulse shaping, are not treated. The assumption also allows us to derive the $\gamma_j$ of each element independently ($\gamma_j(\omega)$ is referred to hereafter as $\gamma_j$).

For homogeneously broadened dyes $\gamma_g$ and $\gamma_a$ are represented as

$$\gamma_g = \frac{\alpha_g + i d_g A_g}{\tilde{E}(\omega)} \int_{-\infty}^{\infty} dt \tilde{E}(t) \exp \left[ - \frac{\Gamma(t)}{\Gamma_g} - i \Delta \omega t \right],$$

$$\gamma_a = -\frac{\alpha_a - i d_a A_a}{\tilde{E}(\omega)} \int_{-\infty}^{\infty} dt \tilde{E}(t) \exp \left[ - \frac{\Gamma(t)}{\Gamma_a} - i \Delta \omega t \right],$$

where

$$\Gamma(t) = \int_{t_0}^{t} dt \Gamma(t')$$

is the cumulative energy in the pulse with the intensity $I(t)$ ($= |E(t)|^2$) and the total energy $\Gamma = \Gamma(\infty)$. The detunings $d_g$ (or $d_a$) are defined as $d_{g(a)} = 2(\omega_{g(a)} - \omega)/\Delta \omega_{g(a)}$, where $\omega_{g(a)}$ and $\Delta \omega_{g(a)}$ are the center wavelength and FWHM of the gain (or the absorption) band. On the assumption of sufficiently broad gain and absorption bands ($\Delta \omega_g > 1/\tau_c$, $\alpha_g$ and $\alpha_a$ are the amplitude gain and the amplitude absorption at the frequency $\omega_0$ before arrival of the pulse, respectively. In addition, $\Gamma_g$ and $\Gamma_a$ are the saturation energies of the gain and absorption media.

The phase retardation by the weak fast SPM is

$$\phi_f(t) = -n_3 I(t) \omega_0 \Delta l/c$$

with the nonlinear refractive index $n_3$ and the medium thickness $\Delta l$. Therefore $\gamma_f$ is represented as

$$\gamma_f = \frac{-i \kappa}{\tilde{E}(\omega)} \int_{-\infty}^{\infty} dt E(t) I(t) \exp(-i \Delta \omega t),$$

where $\kappa = n_3 \omega_0 g \Delta l / c$.

We describe $\gamma_j = (\gamma' + i \gamma'')$ in terms of the amplitude transmission coefficients $\Sigma_i t_i$ and the sum of their dispersions $\Sigma_i \phi_i$, and the prism sequence dispersion $\varphi_p$ as

$$\gamma' = -\sum_i t_i, \quad \gamma'' = \varphi_p + \sum_i \phi_i.$$
The solid curves in Fig. 6 show the \( \varphi^{(2)} \) dependence of \( \tau \) [Eq. (12)] for the present CPM dye laser, where we estimate \( \omega_0 = 2.98 \times 10^{15} \text{ rad/s} \), \( \Gamma_0/\Gamma_s = 6.4 \), \( \Gamma_0/\Gamma_0 = 0.8 \), \( \alpha_0 = 0.041 \), \( \alpha_a = 0.016 \), \( \gamma^{(1)} = -0.025 \), and \( \gamma^{(2)} = -30.0 \text{ fs}^2 \). The CPM effect is taken into account in the estimation of \( \Gamma_0 \). We obtain \( p = -72 \) and \( q = 41 \) from Eq. (13), since \( \omega_a = 3.20 \times 10^{15} \text{ rad/s} \) and \( \omega_g = 3.32 \times 10^{15} \text{ rad/s} \) with, from Ref. 13, \( \Delta \omega_a = 0.22 \times 10^{15} \text{ rad/s} \) and \( \Delta \omega_g = 0.24 \times 10^{15} \text{ rad/s} \) for Rh6G and DODCI. For curves A, B, and C the fast-SPM coefficients \( \kappa \) are equal to 0, 0.4, and 0.8 fs, respectively. The filled circles on the curves indicate the nonchirped pulses \( (c_2 = 0) \).

To estimate the range of \( c_2 \) for which the model of Eq. (12) is reasonable, the numerical results of the frequency-domain approach for a typical non-weak-chirp pulse, \(^{14}\) \( \text{exp}[i \Phi(t/T)] = [\text{sech}(t/T)^{-1}]^2 \) in Eq. (1), are also shown in Fig. 6 as the dashed curves. The curves are obtained by a complete calculation of \( \Delta \omega_p \) terms in Eqs. (3) and (4) with assumptions of constant \( \Gamma_0 \) and \( \omega_0 \). When \( \kappa \tau = 0 \), the solid curves of Eq. (12) almost agree with the dashed curves when \( |c_2/2| \leq 0.4 \). The minimum pulse duration is found on the dashed curve, which is consistent with the usual chirp-compensation model.\(^{1} \) It should be noted that the shape of the dashed curve for positive \( \varphi^{(2)} \) (with large \( c_2 \)) will be influenced more sensitively than that for negative \( \varphi^{(2)} \) by the neglected chirp-dependent frequency shift.

The direction of the parabola in the \( \varphi^{(2)}, \tau \) plane is determined by the sign of \( p \). When \( \omega_0 \) is on the blue side of \( \omega_a \), \( p \) is positive. Then weakly chirped pulses should exist under positive \( \varphi^{(2)} \) (negative GVD). This fact is in agreement with the analysis in the time domain of Ref. 18. In addition, from Eqs. (8) and (9), \( \tau = (-\gamma^{(2)}/R_0)^{1/2} \) when \( c_2 = 0 \). This means that the duration of the pulse without chirp is independent of the weak fast-SPM \( \kappa \), which was first pointed out by Haus and Silberberg.\(^{15} \)

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C. Third-Order-Dispersion Dependence of Pulse Duration

To evaluate the TOD dependence, Eqs. (8)-(11) must be solved simultaneously. The parabolic chirp \( c_2 \) and the optimum second-order dispersion \( \varphi^{(2)} \) are determined in addition to the linear chirp \( c_2 \) and the pulse duration \( \tau \). The derived \( \varphi^{(2)} \) corresponds to the intracavity GVD that is adjusted to produce the shortest \( \text{sech}^2 \) pulse at a given TOD in the experiment.

We treat a case of the negligible fast SPM (\( \kappa = 0 \)). Furthermore \( \gamma^{(3)}/\tau^3 > 0 \) is assumed because the relation \( |\tau_0| > 0 > |\gamma^{(3)}/\tau| \) is satisfied in Eq. (10) for the present laser. With these approximations, the \( \varphi^{(2)} \) dependence of \( \tau \) is written in a simple equation as

\[
\tau^3 + r \tau + s \varphi^{(3)} = 0,
\]

where

\[
r = \begin{bmatrix} T_2 & T_3 \\ U_2 & U_3 \end{bmatrix}, \quad s = \begin{bmatrix} R_2 & R_3 \\ T_2 & T_3 \end{bmatrix}.
\]

When \( \gamma^{(3)}(-\gamma^{(2)}) < 0 \), \( r > 0 \), and \( s > 0 \), Eq. (14) generates the solid curve in Fig. 7. The curve reaches to \( \tau_m = \tau_0/\sqrt{3} \) at the maximum of \( \varphi^{(3)} = 2\tau_0^3/3\sqrt{3}s \), where \( \tau_0 = (-\gamma^{(3)}/\tau^3) \) is the pulse duration at zero TOD \( \varphi^{(3)} = 0 \). The curve is meaningful when the weak-chirp approximation is valid. Since the parameters \( r \) and \( s \) strongly depend on the saturation of the absorber \( [F(t)/F_0] \) and the gain medium \( [F_0(t)/F_0] \), the value of the optimum TOD \( \varphi^{(3)} \) changes remarkably because of both saturation depths.

For the present CPM dye laser (for the circles in Fig. 4) we find that \( r = 77.1 \) and \( s = 37.5 \), from Eq. (15). Therefore Eq. (14) predicts that the pulse duration at the TOD of \( \varphi^{(3)} = 1140 \text{ fs}^3 \) will become \( \tau_p = 1.76 \tau_m = 49 \text{ fs} \). The chirp coefficients at the point \( \varphi^{(3)} \) are found by Eqs. (8),(11), and (14) to be \( c_2/2 = -0.37 \) and \( c_2/6 = 0.08 \). The chirp is weak in the range \( \tau \geq \tau_m \). These results are in good agreement not only with the corresponding experimental value of 50 fs at \( \varphi^{(3)} = 1200 \text{ fs}^3 \) but also with the behavior of the TOD dependence given by the open circles.

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Fig. 6. Pulse durations versus GVD, calculated by the frequency-domain model for the present CPM dye laser. In addition to the results obtained for the weak-chirp model of Eq. (2) (solid curves), the results for the usual chirping model (see text; dashed curves) with various fast SPM's and \( \kappa \) equal to 0, 0.4, and 0.8 fs are shown as curves A, B, and C, respectively.

Fig. 7. Theoretical curves for pulse durations versus TOD for positive \( r \) and \( s \). The normalization factors \( \tau_0 \) and \( \varphi^{(3)} \) are given in the text.
in Fig. 4 in the TOD region smaller than \( \varphi^0 \). This means that, in the larger region with no \( \tau \) solution in Eq. (14), a weakly chirped sech\(^2\)-shaped pulse does not exist, but a nonsech\(^2\) or strongly chirped pulse does. This pulse corresponds to the increasing tails of the autocorrelation traces in the positive region over \( \varphi^0 \) that were observed during the measurements represented by crosses and filled circles in Fig. 4.

The existence of the nonzero optimum \( \varphi^0 \) is understood physically as follows. The gain and absorption modulation is forced to balance frequency dependently with the output coupling at the steady state, \(-i\omega + \text{Re}[\gamma_\text{a}(\omega)] + \text{Re}[\gamma_\text{a}(\omega)] = 0\). Consequently the corresponding phase delay \( \text{Im}[\gamma_\text{a}(\omega)] + \text{Im}[\gamma_\text{a}(\omega)] = \Sigma \varphi_\text{a}(\omega) + \varphi_\text{w}(\omega) \). Therefore, at the minimum pulse duration \( \tau_m \), the same amount of TOD is needed \[ \varphi^0 = 2 \varphi^0_+ + \varphi^0_+ \neq 0 \].

4. CONCLUSIONS

We have experimentally obtained the pulse duration as a precise function of the TOD \[ \varphi^0 \] in a CPM dye (Rh6G + DODCI) laser at -632 nm for the first time to our knowledge. It has been found that the optimum TOD \[ \varphi^0 \] at the point where the shortest pulse duration \( \tau_m \) is generated is not zero but is positive. This result corresponds to the fact that chirp compensation by the prism sequence usually produces shorter pulses than that by the interferometer, since the former and latter have a positive and a negative \( \varphi^0 \), respectively.

It has been shown that an analysis with a frequency-domain description of the slow saturable dyes (in addition to the usual dispersive elements) quantitatively explains the result of the optimum positive \( \varphi^0 \). The reason that the optimum \( \varphi^0 \) becomes positive is that the frequency-dependent phase delay caused by the absorption saturation (the dynamic dispersion) also becomes frequency dependent. To generate the shortest pulse, the frequency-dependent phase delay has to be compensated for by the externally adjustable dispersion, \( \text{Im}[\gamma_\text{a}(\omega)] + \text{Im}[\gamma_\text{a}(\omega)] = \Sigma \varphi_\text{a}(\omega) + \varphi_\text{w}(\omega) \). Therefore, at the minimum pulse duration \( \tau_m \), the same amount of TOD is needed \[ \varphi^0 = 2 \varphi^0_+ + \varphi^0_+ \neq 0 \].

APPENDIX A

The forms of the coefficients \( P_i, Q_i, R_i, S_i, T_i, \) and \( U_i \) \((i = 1, 2, 3)\) in Eqs. (6)–(11) are given here. They are written as

\[ P_i = P_{ig} - P_{ia}, \quad Q_i = Q_{ig} - Q_{ia}, \quad R_i = R_{ig} - R_{ia}, \]
\[ S_i = S_{ig} - S_{ia}, \quad T_i = T_{ig} - T_{ia}, \quad U_i = U_{ig} - U_{ia} \]

where the subscripts \( g \) and \( a \) denote the gain dye and the absorber, respectively. Introducing \( R^0_g \) and \( R^0_a \) as

\[ R^0_g = \frac{1}{\pi} \int_0^\infty d\eta \eta^\delta(\text{sec} \eta) \exp \left[ -\frac{\Gamma_r}{2\Gamma_g} (1 + \tanh \eta) \right] \]

with the normalized time \( \eta = t/\Gamma_r \), we write the terms for a gain dye in the following equations:

\[ P_{ig} = \alpha_g R^0_g, \quad P_{ia} = -\alpha_g d_g R^0_g \]
\[ P_{ig} = -\alpha_g d_g R^0_g, \quad Q_{ig} = \alpha_g d_g R^0_g \]
\[ Q_{ig} = -\alpha_g d_g R^0_g \]
\[ R_{ig} = -\alpha_g d_g R^0_g \]
\[ S_{ig} = -\alpha_g d_g R^0_g \]
\[ S_{ig} = -\alpha_g d_g R^0_g \]
\[ T_{ig} = -\alpha_g d_g R^0_g \]
\[ T_{ig} = -\alpha_g d_g R^0_g \]
\[ U_{ig} = \alpha_g d_g R^0_g \]
\[ U_{ig} = -\alpha_g d_g R^0_g \]
\[ U_{ig} = -\alpha_g d_g R^0_g \]

The corresponding terms for the absorber, \( P_a, Q_a, R_a, S_a, T_a, \) and \( U_a \), are obtained by replacing the subscript \( g \) with \( a \), \( d_g \), and \( R^0_g \) in the above equations.

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REFERENCES

7. J. A. Valdmanis, R. L. Fork, and J. P. Gordon, "Generation of optical pulses as short as 27 femtoseconds directly from a laser balancing self-phase modulation, group-velocity disper-