Positive and negative chirping of laser pulses shorter than 100 fsec in a saturable absorber

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We present a calculation of the chirp generated in laser pulses shorter than 100 fsec on propagation through a saturable absorber (DODCI in ethylene glycol). The calculation takes into account the absorber saturation and the solvent nonlinear refractive index. At pulse energies greater than 10 nJ the chirp tends to be predominantly positive, and it increases rapidly as the pulse duration becomes shorter than 50 fsec. At pulse energies in the 1–7-nJ range the chirp is mostly negative for pulses longer than 30 fsec.

Recently a great deal of interest has been evident in the frequency sweep that exists in pulses generated from mode-locked dye lasers. This frequency chirp is important because it directly affects the durations of the pulses that can be obtained from these lasers. For lasers emitting pulses in the femtosecond domain, a correct balance between the cavity dispersion and the frequency sweep produced by the interaction of the pulse with the saturable-absorber jet is essential if the shortest possible pulses are to be obtained.[1] Martinez *et al.*[2] have presented a theoretical analysis of such lasers in which the phase modulation of the pulse is assumed to be the result of a positive nonlinear index ($n = n_0 + n_2 I; n_2 > 0$). Other authors have found that the laser pulses had a downchirp,[3] which was attributed mainly to absorber saturation.

In this Letter we present a simple calculation of the frequency sweep imposed on femtosecond-duration pulses interacting with a saturable absorber that includes the effect of self-phase modulation (SPM) due to the nonlinear refractive index of the solvent. Depending on the intensity of the propagating pulse, absorber saturation and SPM in the solvent can combine to produce a chirp of either polarity through the main part of the pulse. Other authors have calculated the contribution to the chirp of weak absorber saturation.[3,4] However, in colliding-pulse mode-locked (CPM) lasers, in which the intracavity pulse energy can exceed 5 nJ, the SPM in the solvent becomes important and must be taken into account. Our calculations show that in the case of a typical CPM laser with Gaussian pulses of about 50-fsec duration, the chirp is dominanted by the solvent nonlinearity when the pulse energy density at the saturable absorber exceeds 10 times the absorber saturation energy density.

Consider a Gaussian pulse

$$I(t) = I_0 \exp[-(4 \ln 2)t^2/\tau^2], \qquad (1)$$

which has a FWHM duration τ and energy $E_0 = (1.06)I_0\tau$, propagating through a length l of a saturable absorber in a solvent. Assuming a Gaussian profile simplifies the calculation, and the conclusions drawn are not substantially different from those obtained when the actual laser pulse shape (sech^2) is considered. The optical absorption coefficient at the center of the absorption line is α_0 under unsaturated conditions. Following de Silvestri *et al.*,[5] we consider the absorption line shape to be a Lorentzian,

$$g(\omega) = 2 \times \left\{\pi\Delta\omega\left[1 + \frac{4(\omega_0 - \omega)^2}{\Delta\omega^2}\right]\right\}^{-1},$$

where ω_0 is the center frequency and $\Delta\omega$ is the FWHM linewidth. The contribution of the resonant transition to the refractive index at frequency ω is given by

$$n_R = \frac{\pi}{2}(\omega_0 - \omega)\frac{c}{\omega_0}\alpha g(\omega). \qquad (2)$$

Here α is the saturated absorption coefficient, which, in the case when the pulse duration is much smaller than the energy-relaxation time of the dye molecules, is given by

$$\alpha = \alpha_0 \exp\left[\frac{-\int_{-\infty}^{t} I(\theta)\mathrm{d}\theta}{E_s}\right], \qquad (3)$$

where E_s is the absorber saturation energy density.[6] We have not included the effect of the nonzero phase-relaxation time, T_2, of the absorber. T_2 has been found to be less than 20 fsec,[7] so our analysis for pulse durations longer than 20 fsec should be correct. Rudolph and Wilhelmi[3] have calculated the chirp due to absorber saturation for low saturation levels and have found that the effect of the nonzero T_2 is to decrease the chirp caused by the absorber.

The solvent nonlinearity is described by a term $n_s = n_2 I$ that is included in the refractive index. With this term, the time-dependent phase delay of the pulse on traversing the medium is given by

$$\phi(t) = \frac{-n_2\omega l}{c} I(t) - G(\omega)\frac{\omega l}{c}\alpha(t), \qquad (4)$$

where we have used

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Table 1. Parameters Used for the Calculation of the Frequency Shift $\delta\omega$

Laser wavelength	$\lambda_0 = 610$ nm
Ethylene glycol nonlinear index	$n_2 = 3.0 \times 10^{-16}$ cm²/W
Dye-jet thickness	$l = 50\ \mu$m
Center frequency of DODCI absorption	$\omega_0 = 3.25 \times 10^{15}$ rad/sec
FWHM of the absorption line	$\Delta\omega = 0.22 \times 10^{15}$ rad/sec
Unsaturated absorption at line center	$\alpha_0 = 0.4$
Saturation energy density at 610 nm	$E_s = 0.5$ nJ/cm²
Beam radius at the dye jet	$r = 5\ \mu$m

$$G(\omega) \equiv \frac{\pi}{2}(\omega_0 - \omega)\frac{c}{\omega_0}g(\omega).$$

It is not necessary to include the effects of the absorber on $I(t)$ because typical values for the absorption of an intracavity DODCI jet at 610 nm are of the order of 3%. The same argument stands for the change in the input pulse shape that is due to dispersion since, given a 100-μm ethylene glycol jet, an input pulse of 50 fsec will undergo a variation in its duration of about 10^{-2}%.[5]

The frequency shift $\delta\omega$ induced by the nonlinearities is obtained from the derivative $\delta\omega = \mathrm{d}\phi/\mathrm{d}t$ as

$$\delta w = \frac{-n_2\omega l}{c}\frac{\mathrm{d}I}{\mathrm{d}t} - G(\omega)\frac{\omega l}{c}\frac{\mathrm{d}\alpha(t)}{\mathrm{d}t}\,. \tag{5}$$

For the pulse described by Eq. (1) this shift is

$$\delta\omega_g = \frac{2\pi l}{\lambda}\left\{\frac{5.23 n_2 E_0}{\tau^3}\,t\,\exp\left[-(4\ln 2)\frac{t^2}{\tau^2}\right] + \frac{0.94\alpha_0 E_0}{E_s\tau}G(\omega)\exp\left[-(4\ln 2)\frac{t^2}{\tau^2} - \frac{E_t}{E_s}\right]\right\}, \tag{6}$$

where $E_t \equiv \int_{-\infty}^{t} I(\theta)\mathrm{d}\theta$ and the subscript g in $\delta\omega_g$ indicates a Gaussian input pulse. The values of the parameters used in the calculations are listed in Table 1. The saturation energy density of DODCI has been taken as one third of its true value to compensate for the effect of the two counterpropagating pulses in the absorber jet.[8]

In Fig. 1 we show the instantaneous frequency shift for a 50-fsec pulse at energies of 1 and 15 nJ. These correspond to saturation parameters $\gamma = E_0/E_s$ of 2.55 and 38.1, respectively. It can be seen that as the saturation parameter increases, the chirp, $C = \mathrm{d}^2\phi/\mathrm{d}t^2$, through the main part of the pulse changes from negative to positive. As suggested recently,[1,9] the negative contribution due to the absorber saturation shifts toward the beginning of the pulse as the saturation parameter increases. In addition, the contribution due to n_2 increases with the pulse energy, causing the change in the sign of the chirp at the pulse center that occurs in this case when the pulse energy exceeds about 5 nJ.

To characterize the induced chirp quantitatively, we define an average chirp coefficient, $\bar{C}$, given by

$$\bar{C} = \frac{\int_{-\infty}^{\infty} C(t)I(t)\mathrm{d}t}{\int_{-\infty}^{\infty} I(t)\mathrm{d}t}\,. \tag{7}$$

The sign of $\bar{C}$ gives a fair indication of the predominant chirp in the pulse. For example, in the cases shown in Fig. 1 with pulse energies of 1 and 15 nJ we calculate $\bar{C}$ equal to -0.16×10^{-4}/fsec^{-2} and 0.26×10^{-4}/fsec^{-2}, respectively. In Fig. 2 we show the dependence of this average chirp coefficient on the pulse energy for different values of pulse duration. For 50-fsec pulses the averaged chirp becomes positive at energies above 8 nJ ($\gamma \gtrsim 20$). Increasing the pulse duration causes the chirp to become negative over a broader range of pulse energies. This is because the contribution to $\delta\omega$ of the absorber saturation [the last term in Eq. (6)] scales as $1/\tau$, whereas the contribution

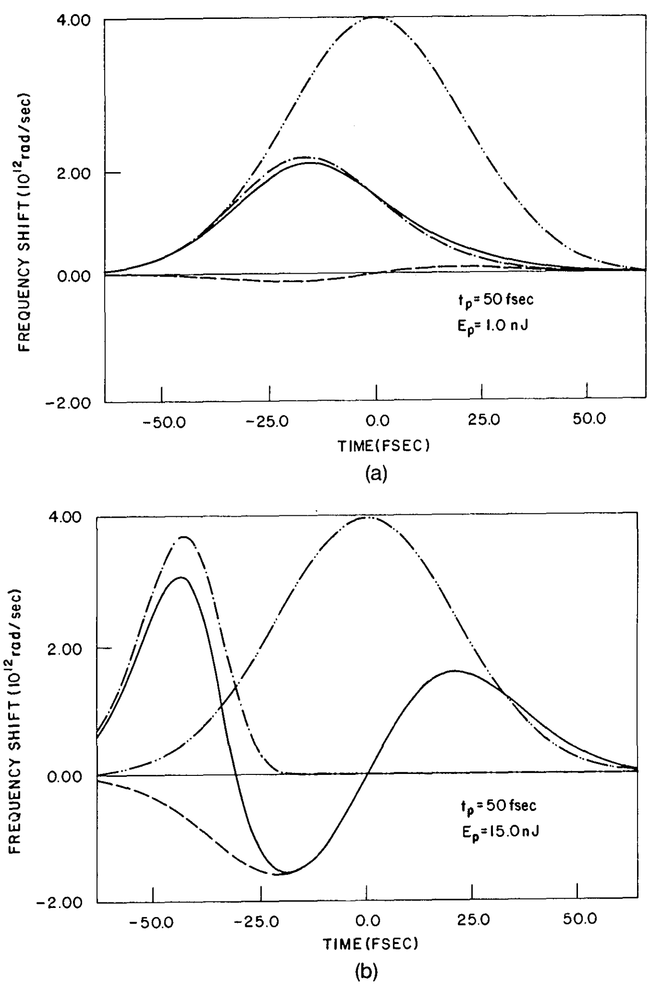

Fig. 1. Instantaneous frequency shift along the pulse caused by the saturation of the absorber (dashed–dotted lines) and by the nonlinear index of the solvent (dashed lines). The resulting shift is shown by the solid line. The input pulse profile is shown for reference (double-dotted–dashed line). The pulse energy is 1.0 nJ in (a) and 15.0 nJ in (b), and the pulse duration is 50 fsec in both cases.

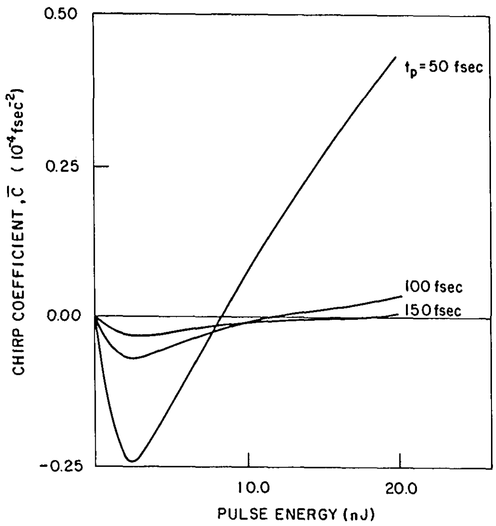

Fig. 2. Average chirp coefficient as a function of the pulse energy for three different values of the pulse duration.

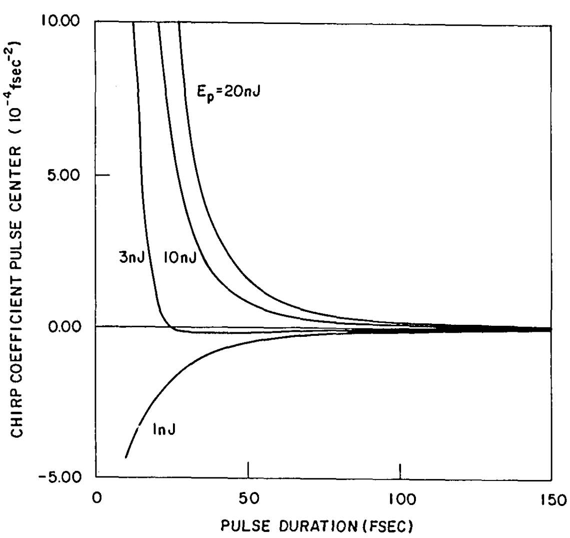

Fig. 3. Chirp coefficient at the peak of the pulse as a function of the pulse duration for four different values of the pulse energy.

of n_2 (fast self-phase modulation) scales as $1/\tau^3$. For the pulse durations shown it can be seen that the negative chirp has a maximum at a pulse energy of about 2.0 nJ ($\gamma = 5.1$). For pulses shorter than 100 fsec, pulse energies in the 10–20-nJ range yield a positive chirp. This can be compensated for within the laser cavity by including an element with negative group-velocity dispersion, such as a four-prism set.[1,10] In the case of lower-energy pulses (1 to 7 nJ), compensation for the negative chirp has been obtained by using an intracavity prism with positive group-velocity dispersion.[11] In Fig. 3 we show the variation of the chirp coefficient at the pulse center, $C(0)$, with the pulse duration. Here it is interesting to note the rapid increase in the positive chirp for 10- and 20-nJ pulses as the pulse duration becomes shorter than 50 fsec. These data can be used to estimate the group-velocity dispersion necessary to compensate for the chirp generated in the saturable-absorber jet to obtain further pulse compression. For example, from Fig. 3 we see that for a 30-fsec, 10-nJ pulse (approximately the situation described in Ref. 1), the chirp at pulse center is $C(0) = 4.0 \times 10^{-4}/\text{fsec}^{-2}$. This corresponds to a frequency shift of $\delta\omega \cong C(0)\tau = 1.2 \times 10^{13}$ rad/sec. The dispersion necessary to compensate for this frequency sweep and compress the pulse to its minimum duration is[12] $d^2P/d\lambda^2 = -1.9 \times 10^8\ \text{m}^{-1}$, where P is the optical path. If a four-prism compensator is used, the required prism spacing is 16 cm.[10]

In conclusion, we have presented a simplified calculation of the chirp generatead in an ultrashort pulse on propagating through a saturable absorber. Depending on the pulse energy and duration, this chirp may be predominantly negative or positive. For pulses of less than 70 fsec and more than 10 nJ the chirp is positive. In this case, an element with negative group-velocity dispersion is required in the laser cavity for shorter pulses to be achieved.

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