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Pulse propagation near zero group-velocity dispersion in a femtosecond dye laser

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Femtosecond pulses have been obtained by using different types of laser, resulting in pulses shorter than 30 fsec. Among these techniques, one of the simplest and most powerful is the colliding-pulse mode-locked (CPM) laser. Since its introduction in 1981, this method has significantly evolved. It became obvious that pulses were formed not only by gain and loss saturation but also as a result of nonlinear effects. It was shown that the shortest pulses are obtained when the intracavity group-velocity dispersion (GVD) and self-phase modulation (SPM) are balanced. This led to the concept of soliton shaping. The experimental proof of the pulses' solitonlike character was obtained recently. In order to obtain this solitonlike propagation, the cavity GVD has to be perfectly controlled. Several authors have studied both experimentally and theoretically the influence of GVD on the laser characteristics. By comparing the results of a numerical model and the autocorrelation functions of the pulses produced by a spectrally windowed CPM laser, Avramopoulos et al. have shown that the shortest pulses are obtained for small negative GVD. Recently we experimentally measured the GVD of our CPM laser by using \( N = 2 \) solitonlike pulse properties and found that the optimum GVD is negative and extremely small. All these results show that the higher-order dispersion terms have to be taken into account. Recently de Barros et al. showed theoretically that the third-order term could have an influence on the pulse characteristics. In this Letter we report an experimental study on the characteristics of pulses produced by a CPM laser operating near the zero of the second-order dispersion and we show that the experimental results are relatively well explained if one takes the third-order term into account.

The experiment was conducted using a classical six-mirror CPM laser including four silica prisms. The dyes used were Rhodamine 6G and DODCI dissolved in ethylene glycol. All the mirrors were single stack and coated for 100% reflectivity at normal incidence at 620 nm. The output coupler had a transmission of 1%. Starting from the alignment that gives the shortest pulses (typically 60 fsec, although we have obtained 31-fsec pulses with this cavity), we increased the DODCI jet thickness by translating the jet vertically. The estimated thickness is then above 50 \( \mu m \). The pulse width increased to approximately 80 fsec, while the wavelength shifted to the red. The exact effect of translating the absorber jet is not perfectly understood. It could increase the effect of SPM and reduce the shaping mechanism due to the absorber and gain. When one of the prisms is translated, introducing a little more glass in the cavity than is needed to get the shortest pulses, a complicated spectral and temporal behavior is observed. A second peak appears in the pulse spectrum at a wavelength that depends on the intracavity GVD. When the spectrum presents two peaks the autocorrelation trace evolves from the usual clean near-sech\(^2\) shape to a complicated multiple-peaked shape depending on the orientation of the autocorrelator doubling crystal (i.e., on the wavelength of the doubled signal). The KDP doubling crystal was 100 \( \mu m \) thick, and its acceptance bandwidth was much narrower than the 20-nm bandwidth of the pulses. We found that the main peak of the spectrum corresponded to a stable and clean pulse. Figure 1 shows the evolution of the pulse spectrum as a function of intracavity GVD variation. A second spectral peak is clearly visible on the right, and its position changes until it merges with the principal part of the spectrum. No modulation of the pulse train envelope reminiscent of high-order solitonlike pulses was observed until the two peaks merged. Figure 2 gives the position of the two peaks as a function of the prism position. Obviously this position cannot be accurately measured for the first curve. When more than two spectral peaks can be seen in Fig. 1, only the two wavelengths with the highest peaks have been considered. Such a splitting of a pulse spectrum has already been observed in an optical fiber and in a CPM laser. It has been attributed to the third-order dispersion when the pulse propagates near the zero second-order dispersion of the fiber or the laser and has been studied by several authors.

When a pulse is launched near zero dispersion, a part of its spectrum lies in the positive dispersion...
obtained in a simple manner. Let us write the group velocity \( V_0 \) at a frequency \( \omega_0 \),

\[
V_0 = \left[ \frac{d}{d\omega} \right]_{\omega=\omega_0}^{-1} k_0
\]

where \( k \) is the wave number. We can obtain the group velocity \( V \) at a wavelength \( \omega \) near \( \omega_0 \) by using a second-order expansion of the group velocity,

\[
V = V_0 + \frac{dV}{d\omega} \bigg|_{\omega=\omega_0} (\omega - \omega_0) + \frac{1}{2} \frac{d^2V}{d\omega^2} \bigg|_{\omega=\omega_0} (\omega - \omega_0)^2.
\]

Introducing the second- and third-order GVD,

\[
k_0'' = \frac{d^2k}{d\omega^2} \bigg|_{\omega=\omega_0} \quad \text{and} \quad k_0''' = \frac{d^3k}{d\omega^3} \bigg|_{\omega=\omega_0}
\]

at the wavelength \( \omega_0 \) and assuming that \( k'' \ll k''' \), we obtain

\[
V = V_0 - \frac{k_0''}{k_0^2} (\omega - \omega_0) - \frac{1}{2} \frac{k_0'''}{k_0^2} (\omega - \omega_0)^2.
\]

We can now look for two wavelengths \( \omega_1 \) and \( \omega_2 \) that have the same velocity. By using Eq. (3), this leads to

\[
(\omega_1 - \omega_2) [k_0''' + \frac{1}{2}k_0''''] (\omega_1 + \omega_2 - 2\omega_0) = 0.
\]

In order to check the fit between Eq. (4) and the experiment, we have plotted in Fig. 3 the variation of the quantity \( \omega_1 + \omega_2 - 2\omega_0 \) obtained from Fig. 2 versus the variation of \( k_0'' \). The frequency \( \omega_0 \) was arbitrarily chosen as the mid-frequency between the two left-most points of Fig. 2. The experimental points are close to the straight line predicted by Eq. (4). A least-squares fit gives a slope of \(-1800 \text{ fsec}^3\), leading to a third-order intracavity dispersion of \(-3600 \text{ fsec}^3\). We note that this value is negative as expected if the major source of third-order dispersion comes from the prisms. We can also use Eq. (4) to estimate the second-order dispersion of the cavity. Using the spectral shift \( \omega_1 + \omega_2 - 2\omega_0 \) corresponding to the top curve of Fig. 1, we obtained \( k_0'''' = -60 \text{ fsec}^2 \) for this particular prism position. In a recent experiment we obtained

Assuming that, in order to be stable, the different parts of the spectrum have to travel at the same speed, the exact position of the spectral resonance can be
The result is approximately -50 fsec$^2$. This value is corresponding to a 50-fsec transform-limited pulse. We have found that the minimal pulse duration achieved in a CPM laser is not currently limited by the laser bandwidth but by high-order dispersion effects.

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