# Nonreciprocal phase shift in a femtosecond dye laser

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Beating is observed when the two output beams from a colliding-pulse mode-locked laser are recombined outside the cavity. This beating is attributed to nonlinear dephasing in the saturable absorber. An explanation in terms of the soliton period variation is given. The experimental results show good agreement with the predictions of the nonlinear Schrödinger equation.

The production of femtosecond pulses in colliding-pulse mode-locked (CPM) dye lasers has been widely studied for several years.[1-4] From these studies it has been established that nonlinear effects govern the pulse evolution[2-6] and that the pulses benefit from soliton shaping owing to the compensation or at least the interaction of intracavity group-velocity dispersion with nonlinear phase modulation. In a simplified approach pulses can be compared with real solitons. The nonlinear Schrödinger equation governing soliton behavior has been shown to describe certain traits of the CPM method.[7-9] We present here some results on phase properties of pulses produced by a CPM laser and show that these results can be explained by using the nonlinear Schrödinger equation.

The experiments were conducted with a classical CPM ring laser (Fig. 1). This laser includes two dye jets (Rhodamine 6G as the amplifier medium, DODCI as the saturable absorber). The pump power is provided by a cw argon-ion laser working at 514.5 nm. The intracavity group-velocity dispersion was controlled by a sequence of four silica prisms.[10] This laser provides two output beams with powers of ~20 mW. The pulse duration was typically 80–100 fsec. First, we studied the jitter between the pulses coming from the two output beams. The two pulses counterpropagating in the laser collide in the saturable absorber but are not in temporal coincidence at the output coupler (OC). A delay line was placed in the beam coming directly from the Rhodamine jet (beam A in Fig. 1). The two beams were recombined with a beam splitter (BS). One output of the beam splitter was sent to the autocorrelator, and the other output was incident upon a slow photodiode.

When the delay between pulse A and pulse B was adjusted to be small but not zero (~200 fsec), a triple-humped correlation trace was observed. The central peak corresponds to the sum of the autocorrelation traces of each pulse, while the two other peaks correspond to intercorrelations between the two pulses. Within our experimental precision, no difference between the shapes of autocorrelation and intercorrelations was observed. We then estimated the jitter between the two counterpropagating pulses to be less than 5 fsec.

We then adjusted the delay line in order to obtain a perfect temporal coincidence of the two pulses on the beam splitter. The autocorrelation and intercorrelations merged, but the trace had a deeply modulated shape. Looking at the pulse train envelope with a slow photodiode, we observed a similar modulation at a frequency of ~10 kHz (Fig. 2). This beat note is attributed to a relative frequency shift of the two pulses. One can see that the modulation is perfectly stable despite the absence of any stabilization system in the laser. The background comes from the imperfect balance between the two pulse energies and can be canceled by using a neutral-density filter on the most powerful beam. The modulation frequency was found to be independent of the position of the prisms in the laser cavity but was related to the pump power and to the position of the DODCI jet relative to the beam waist. The beating frequency varied from 4 to more than 20 kHz when the DODCI jet was translated in the focus region. To determine which of the two pulses had a higher frequency, we applied a translation motion to the mirrors used in the delay line (Fig. 1). This motion introduced a Doppler shift on the pulse frequency. By comparing the direction of the motion and the increase or decrease of the beating frequency, we were able to determine the sign of the dephasing between the two pulses. We found that pulse A always had the higher frequency. We then measured the average power of each beam at the output of the laser and found that the two beams did not have the same power. This power difference was also dependent on DODCI jet position and pump power.
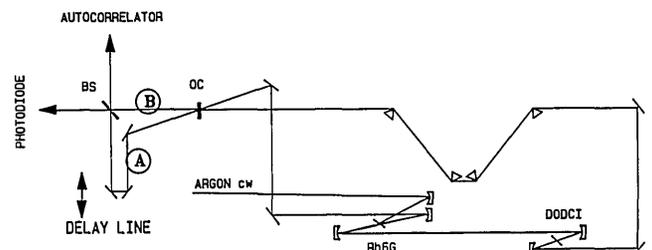


Fig. 1. Experimental setup. The laser cavity is on the right.

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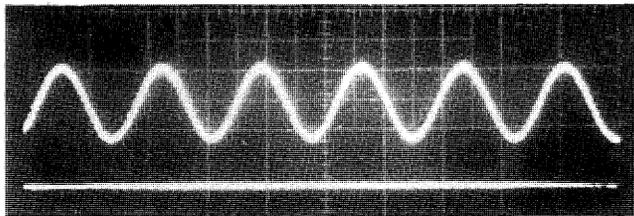
Fig. 2. Modulation of the pulse-train envelope seen by a slow photodiode. The time scale is 20 $\mu$sec per division. The straight line indicates the zero level.
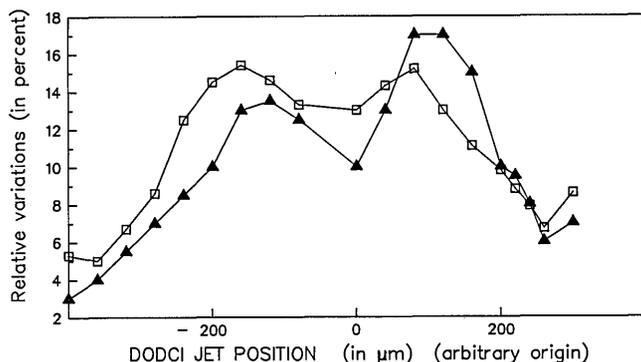


Fig. 3. Evolution of the frequency difference relative to a 100-kHz characteristic frequency (triangles) and of the relative difference of the two output beams' averaged power (squares) versus the position of the DODCI jet relative to the beam waist.

We thus recorded the relative variations of the beams' average power difference $(P_A - P_B)/P_A$ and of their relative frequency difference $(f_A - f_B)/f_0$, where $f_0$ is a 100-kHz characteristic frequency (see below), as a function of the DODCI jet position (Fig. 3). This figure clearly indicates that these two values are correlated. Similar correlations are obtained when the pump power is varied. Note that the curves in Fig. 3 show two maxima that do not correspond to the focus point. It seems likely that the maximum nonlinear effect was not obtained at focus. Such behavior was predicted by Kühlke et al.,[11] and in most of the CPM lasers the shortest pulses are not obtained at the focus point.

We think that the surprising correlation observed in Fig. 3 can be explained in terms of nonlinear dephasing. The explanation is even simpler if one considers that the two pulses present in the laser cavity are $N = 1$ solitons. From the soliton theory it appears that $N = 1$ solitons present a constant temporal shape and exhibit only a periodical phase shift.[12] The period $Z_0$ of this phase shift is given by the soliton period and can be related to the soliton power $P$ by

$$Z_0 = \frac{\lambda A}{4 n_2 P}, \tag{1}$$

where $n_2$ is the nonlinear index of refraction of the propagation medium, $\lambda$ is the pulse wavelength, and $A$ is the beam area in the nonlinear medium. In the case of a laser this expression can be rewritten to introduce the soliton period $N_0$ in terms of the number of cavity round trips. Introducing the cavity round-trip time $T$ leads one to the soliton frequency $f_0$,

$$f_0 = \frac{1}{N_0 T} = \frac{4 n_2 l P}{\lambda A T}, \tag{2}$$

where $l$ is the length of the nonlinear medium in the cavity. Expression (2) shows that there is a linear relation between the soliton frequency and the soliton power.

Let us now consider two solitons that are coherent but have different powers. In order to remain a soliton, the pulse with the lower power must increase its period. These two solitons are then moving at the same velocity but are progressively dephasing. We have checked this explanation by recording the relative difference of soliton frequency $(f_A - f_B)/f_A$ versus the relative difference of power $(P_A - P_B)/P_A$. The problem is to estimate the soliton frequency $f_A$, since we can only measure the beating frequency. This problem was solved recently by studying $N = 2$ solitons with characteristics similar to those of $N = 1$ solitons.[13] It is easy to record the soliton frequency from high-order solitons that exhibit a periodical evolution of their shape. The problem is to find high-order solitons with the same characteristics as usual $N = 1$ solitons (the same wavelength, the same duration, etc.). We recently observed that our laser can produce $N = 2$ solitons similar to the usual fundamental soliton but still having a small modulation of its shape. The soliton frequency, which does not depend on the soliton order, can be deduced from these measurements and apply to $N = 1$ solitons. This experiment has given a soliton frequency that is ~100 kHz in our laser (approximately 1000 cavity round trips) for an 80-fsec pulse. This frequency is a linear function of the intracavity dispersion, but its evolution with defocusing is not well known. Figure 4 gives the relative variations of the beating frequency versus the relative laser output power variations when different laser parameters were changed. First, the DODCI jet was translated relative to the beam waist. Next the jet was held fixed but the pump power was increased from
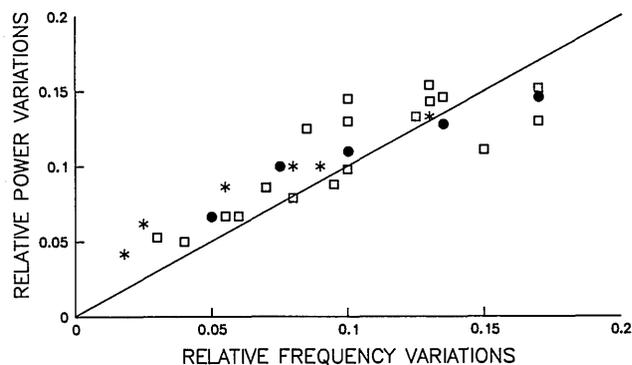


Fig. 4. Relative averaged power difference versus the beating frequency divided by the soliton frequency for changing parameters: DODCI jet position (squares), pump power with the DODCI jet in the first position (asterisks), and pump power with the DODCI jet in the second position (circles). The straight line is the theoretical relation given by expression (2).

1.6 to 1.9 W, for two different positions of the DODCI jet. The straight line is the theoretical relation given by expression (2). The agreement is good and seems to indicate that the beating frequency corresponds to the nonlinear dephasing of the two pulses. According to the soliton theory, these two solitons with different periods should have different durations. Nevertheless the pulse duration is proportional to the square root of the soliton period. This leads to a pulse duration difference of only a few percent and could explain why we did not see a large difference between autocorrelation and cross-correlation functions.

Several questions remain. Among them: why do the pulses have different energies, and how can the pulses stay coherent over more than $10^6$ cavity round trips? We think that the answer to this question arises from the nonsymmetrical structure of our ring laser. As the gain in the Rhodamine 6G jet is saturated, the pulse passing first in the gain medium and then in the lossy section (output coupler and prisms) arrives at the absorber at a lower energy level than the pulse that comes first through the lossy section and then through the gain section. A second point is less obvious. The two pulses encounter noisy media such as dye jets and have no reason to be phase locked. Nevertheless, the pulses are also colliding in the DODCI jet, creating a spatial index grating. This colliding effect has not been taken into account above. It has been shown recently that the influence of this grating depends on the ratio between the pulse duration and the jet thickness.[14] In our laser the jet thickness was over 40 $\mu$m while the pulse duration corresponded to a spatial length of ~20 $\mu$m, leading to a nonlinear dephasing nearly equal to that which would arise for a single pulse passing through the medium. Nevertheless the grating exists and can act as a Bragg reflector, coupling a part of each pulse to the other one. Four-wave mixing in the absorber jet of a femtosecond laser has already been demonstrated,[15] and a nonnegligible reflectivity was observed.[16] This effect could explain the phase coupling between the two pulses, even in presence of noise sources in the laser cavity. However, other mechanisms may also be invoked, and the observed small phase jitter is not well elucidated at present and may also be related to phenomena reported in laser gyroscopes.[17]

In summary, we have observed the nonlinear dephasing of the two pulses counterpropagating in a CPM laser. This dephasing is attributed to the difference of the pulses energies, which leads to a different self-phase modulation through the optical Kerr effect in the DODCI jet. This behavior can also be explained in terms of beating between two solitons that have different energies but the same velocity. The variation law of the beating frequency versus the energy difference is shown to follow the predictions of the nonlinear Schrödinger equation. The beating is stable over more than $10^6$ laser round trips, which reveals a large phase locking between the pulses. This coupling is attributed to four-wave mixing in the DODCI jet. We think that the conjugation of soliton shaping along with phase coupling can explain the good stability observed in CPM lasers.

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