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Characteristics of a femtosecond transform-limited Kerr-lens mode-locked dye laser

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Using an SF6 glass plate as the intracavity Kerr medium and a double-prism pair for dispersion compensation, we developed a femtosecond transform-limited passively mode-locked dye laser. Self-starting mode locking is achieved with a dilute intracavity TCVEBI saturable-absorber jet. Within a 50% power drop the tuning range is 577-606 nm. Pulse characteristics of the laser agree with theoretical predictions based on the Ginzburg-Landau equation.

In the past two years Kerr-lens mode locking (also called self-mode locking) has been demonstrated in various solid-state lasers and proved to be an effective and stable method for generating ultrashort laser pulses. In Kerr-lens mode locking the intracavity self-focusing effect is utilized to produce a bias on the transverse modes of the laser, such that the laser has higher gain for the pulse state than for the cw state. The bias can be produced with an intracavity hard aperture[1,2] or a pump beam that better fits the self-focused lasing beam (soft aperture).[3-5] Because the optical Kerr effect is not sensitive to wavelength, the Kerr-lens mode-locking mechanism does not restrict the wavelength of the laser. It is therefore natural to ask whether the mechanism can be applied to dye lasers, which have quite different characteristics from solid-state lasers yet have been the most widely used tunable lasers for scientific research.

The differences between dye lasers and solid-state lasers that are relevant to Kerr-lens mode locking are as follows: (1) The gain medium of cw dye lasers is usually a thin jet, which is too thin to provide a significant self-focusing effect. Additional intracavity Kerr media must be used. (2) Dynamic gain depletion, which occurs on the time scale of pulse duration, is several orders of magnitude larger in dye lasers than that in solid-state lasers. The effect might work against the self-starting of mode locking and/or the formation of stable steady-state pulses. (3) The intracavity optical power of dye lasers is much smaller than that of solid-state lasers, and the stability is worse; hence it is much more difficult to start the mode-locking process in dye lasers.

In a previous Letter we partially solved the above problems by using CS_2 liquid as the Kerr medium in a dye laser.[6] The optical Kerr effect in CS_2 is 100-fold stronger than that in most solid materials; therefore it compensates well for the low intracavity power of dye lasers. However, because the Kerr-effect response time of CS_2 is as long as 2 ps, the laser produced only picosecond pulses, and the time-bandwidth product was several times the Fourier-transform limit. In this Letter we report our new development in Kerr-lens mode-locked dye lasers. We use SF6 glass as the Kerr medium $(n_2 = 2.3 \times 10^{-19}\ \mathrm{m}^2/\mathrm{W})$,[7] which has practically an instantaneous response time, to obtain femtosecond mode locking. We also use a double-prism sequence to compensate for the large intracavity positive group-velocity dispersion (GVD).[8,9] Finally, we use a less popular but much more durable saturable absorber, TCVEBI,[10] to make the mode locking self-start. The lifetime of TCVEBI is at least several months, in contrast to two weeks for the commonly used DODCI.

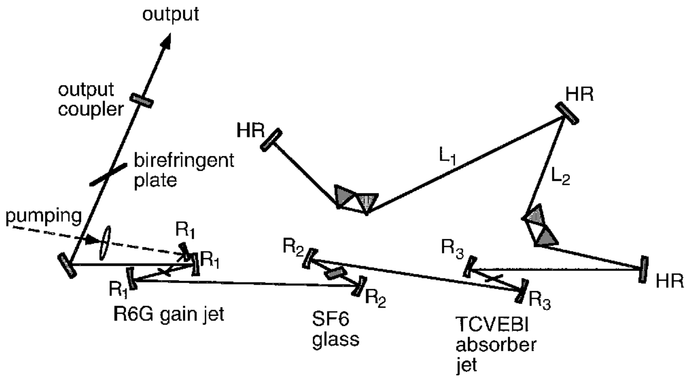

Fig. 1. Schematic diagram of the Kerr-lens mode-locked dye laser. HR's, high reflectors; R_1, R_2, and R_3, concave mirrors with radii of curvature of 10, 7.5, and 5 cm, respectively. $L_1 + L_2 = 61$ cm.

A schematic diagram of the laser is shown in Fig. 1. There are three folded-Z cavities in the laser, one for the 100-μm Rhodamine 590 (R6G) gain jet, one for the 30-μm TCVEBI absorber jet, and one for the 5-mm SF6 glass plate. The cavity round-trip time is 20 ns. The fused-silica double-prism sequence is for producing a large amount of negative GVD to compensate for the positive GVD from the SF6 glass and the birefringent filter. In principle a single-prism sequence with a large prism separation can also provide much negative GVD. In practice, however, a large prism separation makes the cavity inconveniently long and the laser less stable. There are no hard apertures in the cavity to bias the laser against cw operation. We provide effective aperturing by controlling the sizes of the pump beam and the lasing beam and their overlap at the gain jet. To increase the intracavity power, we use an output coupler of 99% reflectivity. The concentration of the gain jet is chosen such that it absorbs 80% of the pump power from an argon-ion laser, and the concentration of the absorber jet is 6.2×10^{-5} M. In both jets the solvent is ethylene glycol. When mode locked, the laser produces 5-mW average power and five well-separated pulses per round-trip time.[11] In the experiments we measure the pulse duration with a real-time background-free autocorrelator and the spectrum with a real-time optical multichannel analyzer. The pulse energy and the envelope of the pulse train are monitored by a fast photodiode.

Kerr-lens mode locking is an efficient mode-locking mechanism because the pulse-shortening effect increases with the peak intensity as the pulse shortens. However, as mode locking starts from intensity fluctuation or mode beating, the peak intensity of the fluctuation is 10,000- to 100,000-fold smaller than that of the steady-state mode-locked pulse. Therefore the pulse-shortening effect in the starting stage is also smaller than that in the steady state by the same proportion. In general, Kerr-lens mode-locked lasers do not self-start with such a small initial pulse-shortening effect. We use a dilute absorber jet to make the mode locking self-start. According to our previous analysis,[12] dilute dye saturable absorbers greatly enhance the pulse-shortening effect in the starting stage because of their large absorption cross sections. The required concentration is so small that the absorber does not have much effect on much the wavelength tuning range. In our laser the absorber produces less than 1% linear loss within the tuning range.

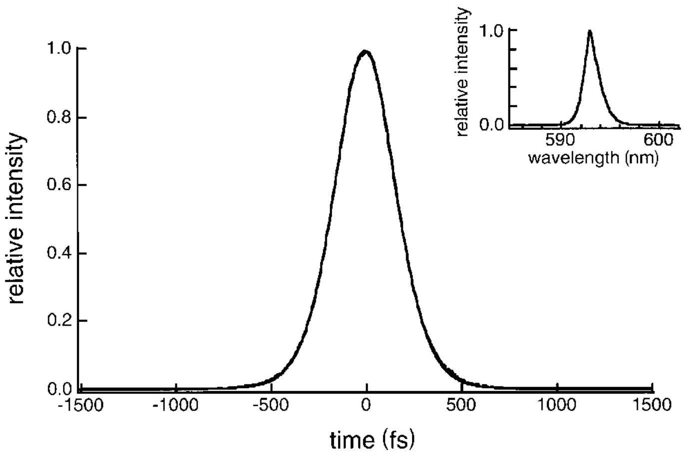

Fig. 2. Autocorrelation trace of the output pulses at 593 nm and the $sech^2$ fitting curve. Inset: the corresponding power spectrum.

A typical real-time autocorrelation trace at 593 nm is shown in Fig. 2. The inset shows the spectrum. The autocorrelation curve fits well that of 240-fs $sech^2$ pulses, as shown by the solid fitting curve, which almost completely merges with the data curve. The time-bandwidth product of the pulses is 1.1 times the Fourier-transform limit. The wavelength of the laser is tuned by a one-plate birefringent filter (Spectra-Physics 0434-8931). For output power within 50% of its peak value, the tuning range is 577-606 nm. Within the tuning range the pulse duration is smaller than 500 fs, and the time-bandwidth product is maintained at $\approx$1.1 times the Fourier-transform limit. We have also mode locked the laser with a ring configuration. However, only picosecond pulses are obtained.

The SF6 glass plate placed at Brewster's angle provides the optical Kerr effect in the cavity. Without the glass plate, only mode beating is observed. Mode locking is sensitive to the following parameters: (1) The position of the glass plate, which determines the power dependence of the lasing mode size and hence affects not only the efficiency of the aperture but also the optimal positions at which to place the aperture. (2) The position of the pump beam lens, which determines the size of the pump beam at the gain jet and hence affects the efficiency of the gain aperture. (3) The distance between the focusing mirrors in the Z folds, which determines the size of the lasing beam and hence also affects the efficiency of the gain aperture. (4) The position of the prism sequence, which determines the amount of GVD. (5) The pump power, which controls the intracavity power. Sensitivity to the first three parameters described above suggests that self-focusing is an important element of the pulse-shortening mechanism. It also suggest that self-phase modulation and GVD *alone* cannot account for the entire pulse-shortening effect, even though the interaction between them is an important pulse-shaping mechanism in femtosecond lasers: if they could account for the entire effect, such sensitivity should not exist. Within the stability region we can control the pulse duration up to a factor of 2 with both the Kerr effect and the GVD while maintaining the near-transform-limit time-bandwidth product. When the Kerr effect is increased by moving the glass plate toward the beam waist, the negative GVD must be decreased (its absolute value increased) to maintain a constant pulse duration. This observed trend agrees with theories based on the Ginzburg-Landau equation.[13]

The laser mode locks stably only at a pump power within $\sim$0.2 W above the lasing threshold. Larger pump power causes microsecond-scale output power fluctuations. Because the relative sizes of the pump beam and the lasing beam affect not only the effectiveness of the gain aperture but also the overall gain, the lasing threshold depends on how far the laser is biased against the cw state. Without removing any of the components for mode locking, when we optimize the laser to produce cw output the threshold is 3.7 W. The threshold increases to 5.4 W when the laser biased for 240-fs pulses. At this point, there is not much room left for further increase of the threshold, because the dye starts to bleach at higher pump power.

To make a quantitative comparison with theories, we calculated the four parameters, gain bandwidth $_g$, GVD

D, self-phase modulation δ, and loss saturation γ, used in the Ginzburg-Landau equation from parameters of our laser. The formula for the transfer function for birefringent filters can be found in Ref. 14, and the GVD for multiprism sequences is given in Ref. 8. Dispersion data of the SF6 glass were obtained from Schott Glass Technologies, Inc. With the above information we calculate $_g$ and D. To calculate the other two parameters, δ and γ, we first measure the size of the lasing beam near the output coupler as well as the size and divergence of the beam reflected from the glass plate, then calculate the beam size at the gain jet and in the SF6 glass. Once the beam size is known, δ can be calculated from n_2 of the glass and γ can be calculated from the procedure described in Ref. 4. With the above parameters we then obtain the pulse duration and the chirp parameter from solutions of the Ginzburg-Landau equation.[13] The values predicted with the Ginzburg-Landau equation agree with our experimental data to within 20%. The small deviation may be due to effects that are not taken into account in the Ginzburg-Landau equation, such as the dynamic gain depletion and its associated nonlinear chirp[15] or higher-order nonlinear gain and GVD. The discrete nature of various components in the cavity is also somewhat different from that of the continuous model described by the Ginzburg-Landau equation.

Figure 3 shows the stability region predicted by the theory in Ref. 13 as well as the operating region of our laser. The agreement is fairly good. The figure also shows that we have pushed the laser close to the stability edge to minimize the pulse duration. By the theory in Ref. 13, there exist stable solutions of the Ginzburg-Landau equation with a pulse duration close to 100 fs and a larger pulse energy. However, with the current design of our laser we are not able to increase the intracavity power further.

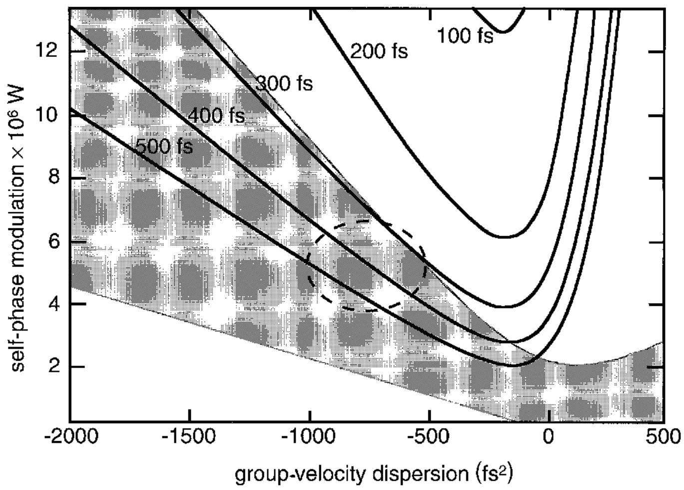

Fig. 3. Theoretical stability region (shaded area). The operating region of the laser is enclosed by the dashed ellipse.

In summary, we have developed a widely tunable, self-starting, femtosecond Kerr-lens mode-locked dye laser. The laser produces near-transform-limit pulses within its tuning range. The characteristics and stability region of the laser agree with theoretical predictions based on the Ginzburg-Landau equation. We also find that dynamic gain depletion, which accounts for the most significant difference between dye lasers and solid-state lasers, does not seem to have negative effects on the characteristics of the laser. Our experiments eliminate such concerns and thereby encourage further developments of Kerr-lens mode locking in other dye-laser systems.

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