Generation of pulses as short as 93 fs from self-starting femtosecond Cr:LiSrAlF₆ lasers by exploiting multiple-quantum-well absorbers

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A Cr:LiSrAlF₆ laser has been mode locked by using a multiple-quantum-well (MQW) absorber. With the MQW absorber inside the laser cavity, Kerr lens mode locking is initiated, yielding transform-limited pulses as short as 93 fs. Pulses of 500-fs duration have also been produced by using resonant passive mode locking with the MQW absorber in an external cavity.

Ultrafast solid-state lasers have been revolutionized in recent years by the demonstration of self-mode locking in titanium-doped sapphire (Ti:Al₂O₃) lasers, whereby pulses of less than 20 fs in duration can now be routinely generated. The process of Kerr lens mode locking (KLM), which is utilized in these systems, is generally not self-starting, and so some additional amplitude modulation is required for initiating the pulse formation, e.g., synchronous pumping, passive mode locking with a fast saturable absorber dye, resonant passive mode locking (RPM) with a semiconductor saturable absorber, or active mode locking with an acousto-optic modulator. Self-mode locking through KLM has also been demonstrated in other established solid-state media such as Nd:YLF (Ref. 9) and Nd:YAG. The laser material Cr³⁺:LiSrAlF₆ (Cr:LiSAF) (Ref. 11) has a bandwidth and tuning range similar to that of Ti:Al₂O₃ and is an attractive alternative for many applications; in particular, it has absorption bands in the red spectral region that allow for diode pumping by the new generation of laser diodes. The advances in Ti:sapphire self-mode-locked lasers have been paralleled by similar progress in Cr:LiSAF lasers. Pulses of 50-fs duration have already been produced by using KLM in Cr:LiSAF with an acousto-optic modulator and a saturable absorber dye as the starting mechanisms. We have also recently demonstrated the generation of pulses of 33 fs in duration from a KLM Cr:LiSAF laser at pump power levels well within reach of currently available diode lasers. This laser used a saturable absorber dye to start the self-mode locking. An all-solid-state self-starting mode-locked femtosecond Cr:LiSAF laser is clearly more attractive for many applications, and so the elimination of the dye jet stream is a necessary step. Since KLM in our Cr:LiSAF laser is not self-starting, we have used a multiple-quantum-well (MQW) device to replace the saturable absorber dye and to initiate mode locking. This MQW absorber was originally developed to act as a saturable absorber in a semiconductor diode-laser system.

As in our earlier studies, the 488-nm line from an argon-ion laser was used to pump a 22-mm-long, Brewster-angled rod of Cr:LiSAF by using an antireflection-coated, 10-cm focal-length lens. The astigmatically compensated laser cavity, as shown in Fig. 1, consisted of two 10-cm radius-of-curvature folding mirrors (M1 and M2), a 1% output coupler (M3), two Brewster-angled SF10 glass prisms (P1 and P2), and the MQW absorber at the focus of a 50-mm radius-of-curvature highly reflecting mirror (M4). A vertical adjustable slit could be inserted between M3 and P2 as required. For all the research reported here, pump powers quoted refer to absorbed pump power, which was 56% of incident pump power (30% was lost through unwanted reflections at M1, and 80% of the remainder was absorbed in the laser rod). The MQW absorber consisted of 100 periods of 7-nm-thick GaAs wells with 10-nm-thick AlGaAs barriers. Proton implantation was used to reduce the carrier lifetime to 150 ps. The excitonic absorption peak was at 830 nm at room temperature. The surface of the absorber had a single-layer antireflection coating and was placed on a silicon wafer that had
been coated with silver. This was mounted on a copper block that was water cooled and maintained at a temperature of ~5°C. A more detailed discussion of the MQW absorber is given in Ref. 16. With no MQW absorber in the cavity, the cw lasing threshold was 160 mW of absorbed pump power. By introducing the MQW absorber assembly to form the high reflector at one end of the cavity, we increased the lasing threshold to 350 mW of absorbed pump power. For pump powers within ~100 mW of threshold, we were able to obtain cw or self-Q-switched operation. At an absorbed pump power of 480 mW, stable self-starting cw mode locking was observed, and self-Q-switching was not a problem. Careful adjustment of the focusing of the gain-folded section and the position of the laser rod resulted in femtosecond pulse generation.

At an apex-to-apex prism separation of 26 cm, pulses of ~90-fs duration (assuming a sech² pulse profile) were measured. The autocorrelation profile of these pulses, together with the associated spectrum, is shown in Fig. 2. Pulse-width measurements were made by using a collinear second-harmonic-generation autocorrelator. The time-bandwidth product for these pulse is ΔνΔτ = 0.33, which is within 5% of the transform-limited value expected for sech² pulses. The output power with the 1% output coupler was 5 mW for 480 mW of absorbed pump power. Whereas this is low compared with that typically obtained with Ti:sapphire lasers, it corresponds to a slope efficiency of approximately half that obtained from our Cr:LiSAF laser without the MQW absorber. The reduction is due partly to the insertion loss of the MQW absorber and partly to the alignment requirements of KLM and of effective saturation of the absorber, which are not those for maximum output power. The introduction of the slit between M3 and P2 stabilized the spectrum and the temporal profile of the mode-locked pulse trains. The use of the intracavity MQW absorber to mode lock the laser always resulted in a shift of the laser spectrum to ~850 nm from its free-running wavelength of ~825 nm. We attribute this to a decrease in the MQW-absorber effective bandgap energy owing to local heating of the absorber by the laser radiation. This laser was not readily tunable, although small changes in alignment and translation of the intracavity slit could shift the center wavelength from ~850 to ~854 nm. In the absence of any dispersion compensation inside the laser, stable cw mode-locked pulses of ~30 ps were obtained at ~850 nm.

Thus the use of the intracavity MQW absorber appeared to provide a sufficiently strong amplitude modulation to initiate Kerr lens mode locking and to produce mode-locked trains of sub-100-fs pulses in a laser that was self-starting. In this system, the sensitivity of the femtosecond laser operation to the precise adjustment of the cavity focusing and to the influence of the intracavity slit suggests that KLM dominated the steady-state laser dynamics, rather than passive mode locking with an ultrafast resonant nonlinear. This would be expected to yield pulses of a few hundred femtoseconds in duration. A possible disadvantage of employing an intracavity MQW absorber is that the minimum achievable pulse duration is likely to be ultimately limited by its frequency response, i.e., by the bandwidth limitation of any étalon structure in the absorber, by the group-velocity dispersion it introduces, and by nonlinear frequency chirp arising from the time-dependent saturation of the absorption.

The MQW absorber was also employed inside an external cavity to investigate the regime of RPM, which has produced picosecond pulses and has initiated KLM in Ti:Al₂O₃ lasers. The laser cavity was modified as shown in Fig. 3 such that M4 was a 3.5% output coupler. The external cavity consisted of two high reflectors (M5 and M6) and the MQW absorber, which was used at normal incidence at the focus of a 20x microscope objective. The length of the main cavity could be adjusted through the position at which it was matched with the external cavity. In RPM lasers there is no requirement actively to maintain a precise match of the main and external cavity lengths, although the pulse duration is sensitive to the mismatch of the cavities. The cw lasing threshold of this cavity was 160 mW of absorbed pump power. At a pump power of 880 mW, ~40 mW was obtained at the output coupler. Stable self-starting cw mode locking was achieved at this pump power, and the shortest pulses obtained were of ~500-fs duration at 842 nm. The shorter operating wavelength, compared to that of the laser with the intracavity absorber, was attributed to a reduced temperature-induced shift of the exciton peak owing to the lower incident average power. Figure 4 shows the autocorrelation profile of these pulses for which the time-averaged spectral width was 3.2 nm. No attempt was made to stabilize the match between the laser and the external cavity lengths, and so this time-averaged spectral width may have represented a narrower optical spectrum fluctuating in frequency. In contrast to the results of Ref. 7, here the mode...
locking was not observed to be self-sustaining if the external cavity was blocked. Also, adjustment of the cavity focusing and the insertion of a slit in the laser cavity adjacent to mirror M4 did not affect the mode-locked performance in the way that would be expected if KLM were important in this laser. An increase in the mismatch of the two cavities resulted in an increase in the pulse duration, as has been observed by Keller et al.\textsuperscript{19} For mismatches of greater than \(-0.5\) mm, only picosecond pulses were produced, and the mode locking became highly unstable. We conclude that the mode-locking dynamics appeared to fit the RPM model for which saturable absorption of the MQW device in the external cavity is the dominant process rather than KLM in the main cavity. For this laser, it was necessary to focus all the output power, which was approximately ten times lower than that of the Ti:Al\(_2\)O\(_3\) laser described in Ref. 7, onto the MQW absorber to achieve saturation, and this entailed maintaining a relatively strong coupling to the main laser cavity. As discussed in Ref. 7, for such strong coupling the MQW-absorber RPM can interfere with the KLM process, and this may explain why we did not observe KLM to dominate the mode locking and generate shorter pulses from this laser.

In conclusion, we have used a MQW absorber inside a Cr:LiSIASF laser to initiate KLM and have generated stable self-starting pulses of \(-90\) fs in duration. We have also utilized RPM with a similar laser and produced stable self-starting pulses of \(500\) fs in duration. It may be possible to refine the laser further and initiate KLM by using a MQW absorber in an external cavity, so that the tuning range and ultimate performance of the laser will not be so compromised by the MQW absorber. For the case of a diode-pumped Cr:LiSIASF laser, however, where the output power is likely to be considerably less than that available from a Ti:Al\(_2\)O\(_3\) laser, taking advantage of the higher intracavity powers available may prove the best approach in the construction of a compact diode-pumped all-solid-state femtosecond laser. One possible improvement might be to use an antiresonant Fabry–Perot saturable absorber\textsuperscript{20,21} either for RPM operation or to initiate KLM. It has been proposed to initiate KLM in a Ti:Al\(_2\)O\(_3\) laser by using this saturable absorber.\textsuperscript{21} The tunability of the laser is clearly compromised by the MQW absorber. This drawback may be partially overcome, however, by band-gap engineering of the MQW material to achieve the desired laser wavelength, as was pointed out in Ref. 20. Whereas an argon-ion-pumped Ti:Al\(_2\)O\(_3\) laser remains the most attractive scientific laser for many applications, an all-solid-state Cr:LiSIASF laser will be an attractive low-cost low-power alternative, and frequency doubling should lead to a compact diode-pumped blue laser source.

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