Stable multipulse generation from a self-mode-locked Ti:sapphire laser

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Abstract

Multipulses up to 6 pulses are generated from a self-mode-locked Ti:sapphire laser with slightly altered cavity configurations. Double and triple pulse operations are found to be very stable and last for several hours. It is noted that the pulse interval takes discrete values of ~ 80 fs interval and its distribution is strongly dependent on the stability of the multipulse oscillation. The laser spectrum in multipulse operation shows a clear periodic modulation, indicating that pulse shaping is attained under the transform-limited condition. It is demonstrated that the application of the external pulse shaping technique to the present multipulse generation widely expands the pulse shaping capability. © 1998 Elsevier Science B.V. All rights reserved.

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The generation of a pulse train in the femtosecond regime has attracted a great interest in the field of optical communications and also in the field of material science such as coherent phonon generation [1]. Among these, phase and/or amplitude modulation techniques for Fourier-transformed femtosecond light pulses are widely used since the development by Weiner et al. [2], and various types of modulation technique have been introduced [3,4]. The advantage of this method lies in the fact that, in principle, arbitrary pulse shape or pulse train with controlled amplitude, phase, and polarization can be generated outside the laser cavity. On the other hand, considerable diffraction loss is inevitable owing to the fine spatial modulation and also various optical components which may possess higher-order dispersion of the refractive index are necessary.

Recently we have developed a simple mode-locked Ti:sapphire laser using a thin Ti:sapphire crystal and have obtained the limiting pulse width of 7.7 fs [5]. In the course of exploitation of this laser, we have found that multipulse is very stably generated owing to the slight displacement of the concave mirrors within the cavity. Although rather unstable multipulse generation has been often encountered when the adjustment of this type of laser is carried out, only a few qualitative experiments have been reported up to now [6,7]. Wang et al. [7] reported on double and triple pulse generation from a self-mode-locked Ti:sapphire laser and showed that the pulse separation in double pulse oscillation takes a discrete value of 64–51 fs. This phenomenon seems very interesting particularly to consider the mechanism of multipulse generation and seems to have potential capability to open a new field for external control of multipulse generation within the laser cavity. In the present paper, we will report more detailed investigations on this phenomenon and will show that the pulse interval is not regular but takes definite values once the cavity parameters are determined. Further, it is shown that the distribution of the pulse interval is strongly dependent on the stability of multipulse oscillation. We will also demonstrate a tentative result on the application of the
external pulse shaping technique to the internally modulated light pulse.

In Fig. 1a, we show the block diagram of a self-mode-locked Ti:sapphire (TiS) laser constructed in our laboratory [5]. Briefly, the TiS crystal employed had 2 mm thickness with a section of $1 \times 5$ mm$^2$ and the absorption coefficient of 4.7 cm$^{-1}$ at 514 nm. The cavity design was analogous to that reported by the Washington State University group [8]. Namely, we employed an asymmetric four-mirror cavity with focal lengths of $\propto (M1, M4)$ and 50 mm ($M2, M3$), and mirror-to-mirror distances of 59, 10, and 101 cm for $M1-M2$, $M2-M3$ and $M3-M4$, respectively. Two quartz prisms with apex-to-apex separation of 40 cm were inserted into the cavity to compensate the group velocity dispersion (GVD). The exciting power source was an all-line Ar$^+$ laser (Spectra Physics 2060) which was focused into the laser crystal with a lens of 120 mm focal length.

The beginning of mode-locking was performed by quick pushing and pulling the prism within the cavity. Although we did not employ an aperture to help mode-locking operation, the single-pulse oscillation was found to be very stable under proper adjustment of the cavity. The mode-locking mechanism without an aperture has been discussed recently in terms of the nonlinear diffraction loss [9]. We normally used output couplers of 10% and 3% transmittance. The minimum pulse width of 7.7 fs was attained when the 3% output coupler was employed. Multipulse generation was also observed mainly with the 3% output coupler. The output power of $\approx 400$ mW is normally obtained under 5 W excitation both for a single pulse and multipulse oscillations with the slightly broader pulse width of 14–15 fs using the 3% coupler.

First we investigate the relationship between the cavity configuration and the output pulse shape. From ABCD ray matrix analysis, the stably oscillating region in an asym-

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Fig. 1. (a) Schematic diagram of a self-mode-locked Ti:sapphire laser constructed in our laboratory. The map of the oscillating states with respect to the displacement of the concave mirrors $M2$ and $M3$ is shown in (b). (c) Enlarged view of the region indicated as dashed lines in (b). Open circles: stable single pulse oscillation, triangles: unstable pulse oscillation, crosses: CW oscillation, full circles: stable multipulse oscillation.
metric four-mirror cavity is divided into two regions with respect to the distance between two concave mirrors [5,9–11]. The Kerr-lens mode-locking is then incorporated by introducing the nonlinear self-focusing and self-shortening effects [10]. It is expected that the change in relative spot size at the end mirror becomes maximum at the inner edges of the two oscillating regions with respect to the M2–M3 distance. Accordingly two pulsation regions due to the Kerr lens effect become largely separated. This result is well reproduced by the observed behavior of the pulsation region as shown in Fig. 1b. It is also found that one pulsation region is surrounded by the stable CW oscillating region and between the CW and the single-pulse pulsation regions various types of oscillation state appear.

We have performed a more detailed investigation on the oscillating characteristics of the mode-locking region. It is found that at the center of the mode-locking region, a stable single-pulse oscillation appears, while at the perimeter of the mode-locking region multipulse and unstable mode-locking oscillations take place and sometimes the coexistence of CW and mode-locking oscillations is observed. It is generally seen that the closer the system approaches the CW region, the more frequently multipulse oscillation takes place with increasing number of pulses. Between stable and unstable mode-locking regions, stable multipulse oscillation which consists of a small number of pulses takes place, shown as full circles in Fig. 1c. In contrast, the multipulse of four or more pulses, which is located in the outer region, is inferior in stability and the change in the number of pulses or the interval is frequently observed even during one oscillating state.

The typical intensity autocorrelation traces of the double pulse oscillation are shown in Fig. 2. It is found that a double pulse having almost equal pulse height is very stably generated. It is stable enough to continue for several hours even under slight mechanical disturbance, unless its mode-locking operation is hindered. Once mode-locking is stopped, double pulse oscillation changes into a different state, i.e. change in pulse separation. Wang et al. [6] reported that the pulse separation in double pulse operation changed with a discrete time interval of \( \delta t_{\text{step}} = 64 \pm 2 \text{ fs} \) for a 35 fs pulse and was dependent on the original pulse

Fig. 2. Autocorrelation traces (left) of double pulses for various pulse separations and their spectra (right) measured simultaneously.
width. This observation is partly reproduced by the present experiment. Namely, for a 15 fs pulse, the pulse separation is found to vary stepwise with the interval of ~80 fs and have the minimum value of ~100 fs. However, we have found that the step is not so regular as shown in Figs. 2 and 3. The distribution of the pulse separations sometimes extends over 10 ps but usually lies within 1 ps. The pulse separation and its distribution are dependent on the cavity configuration, but once the cavity parameters are determined, the reproducibility is quite high. We have measured the distribution of the pulse separations according to the following procedure; the mode-locking operation is ceased by inserting an obstruction into the cavity and then the double pulse is generated through the slight mechanical perturbation. Three typical examples for slightly different M2–M3 or TiS–M2 distances are shown in Fig. 3. Figs. 3a and 3b show that the double pulses with the shorter separations are more likely to occur, while in Figs. 3c the distribution is favorable to the longer separations of 1 ps or more. In every case, each pulse separation falls within 2–3 fs accuracy, thus high reproducibility is obtained. In general, when the double pulse oscillates stably, the pulse separation is not so widely distributed and the probability decreases quickly as the separation increases. It is also noticed that the pulse separation is rather independent of the amount of prism insertion and hence of the pulse width.

In triple pulse operation, the pulse train consisting of almost equal intensity pulses is usually generated with a cavity configuration slightly different from the double pulse case. In most cases, the pulse separation is irregular as shown in Figs. 4b and 4c, but sometimes regular pulse separation is seen (Fig. 4a). In all cases, the pulses are stable for hours or more. There is a tendency that the larger the number of pulses, the less the stability. We have noticed that a maximum of 6 pulses can be generated fairly stably as shown in Fig. 4d.

To investigate the spectral properties of multipulse oscillation, we have measured the laser spectra under various multipulse oscillating conditions. The laser spectra show quite regular modulations for double pulse operation as shown in Fig. 2. The modulation period agrees completely with that expected for transform-limited double pulse. It is found that the depth of modulation is dependent on the amount of prism insertion and under the well-adjusted condition, almost complete modulation is attained, which indicates that the transform-limited condition is satisfied. It is also found that during prism insertion the modulation depth and the whole spectral envelope change remarkably but its period is almost unchanged. The spec-

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![Fig. 3](image-url)  
**Fig. 3.** Distributions of pulse separations in the double pulse regime for three different cavity configurations (a)–(c). Each distribution was obtained after 100–300 trials of ceasing and restarting of double pulse operation.
Fig. 4. (a)-(c) Autocorrelation traces and spectra of triple pulses for three different pulse intervals. (d) Autocorrelation trace of a sextuple pulse.

...trum for a triple pulse seems complicated (Fig. 4) but is explained in terms of the overlap of the modulation due to the three different pairs of pulses.

Finally, to pursue the possibility of external modulation to the multipulse thus generated, we applied the external pulse shaping technique with a grating pair and a spatial light modulator. In this experiment, we employed two 300 grooves/mm gold-coated gratings (Shimadzu Corp.) separated by 80 cm. Between the gratings there were two quartz lenses with the focal lengths of 20 cm and a liquid crystal spatial light modulator (CRI SLM-256). First we investigated the temporal properties of the pulse shaping apparatus. For this purpose, we measured the pulse broadening due to the pulse shaping apparatus by introducing short pulses with various temporal widths without applying any spatial modulation. Although slight broadening and deformation of the pulse profile were always observed, we could confirm that this combination was capable of converting the minimum 20 fs light pulse into arbitrary pulse shape without degradation of the temporal properties.

Generally this type of pulse shaping is realized by adding a filter function to the original Fourier-transformed pulse. The pulse shaping is thus characterized by the convolution of the original pulse shape with the inverse Fourier-transform of the filter function. Since the light pulse under the multipulse oscillating condition is expressed by clear sinusoidal amplitude modulation in frequency domain, its effect on the present pulse shaping is expressed as an additional amplitude modulation to the filter function. Thus the amplitude modulation caused by multipulse oscillation and external modulation work independently, and appear as the duplicate convolution in the time domain. To investigate this, the double pulse separated by 259 fs was generated from the laser cavity and then the binary phase modulation of 22 nm wavelength interval, which converted a single pulse into three pulses with nearly equal intensities was applied. Fig. 5a shows the result of this hybrid modulation. The same procedure was applied to the originally 394 fs separated double pulse. By applying 9.8 nm wavelength modulation, the pulse interval was adjusted so that one pulse among three overlapped with each other, resulting in 5 pulses from the double pulse (Fig. 5b). Since the complete amplitude modulations without loss of intensity and beam quality are rather difficult using the above pulse shaping apparatus, the combination of external and internal modulations may offer a new methodology to femtosecond pulse shaping technology.

The origin of multipulse generation has been discussed frequently in the literature [6,7,9,12-15]. Brabec et al. [12] and Spielmann et al. [6] suggested that the stability of a solitary pulse in a self-mode-locked laser is governed by the combined action of self-phase modulation (SPM) and the GVD. When the effect of the SPM is not so large, the small positive chirp caused by the SPM is effectively compensated by the negative GVD produced by the prism pair. However, with increasing SPM, its effect appears as a strongly nonlinear chirp and only the central part of the pulse is compensated by the prism pair, which results in growth of the side-lobe pulses and, as a result, causes the...
pulse to split [6,7]. Self amplitude modulation (SAM) prevents the pulse from splitting by decreasing the cavity loss at the central part of the pulse time profile and stable single pulse operation is realized. Hence, when the SPM becomes dominating over the SAM, the pulse tends to split. The present experimental observation is directly comparable with the Kerr-lens sensitivity calculated recently for an asymmetric cavity without an aperture [9]. Namely, the regions of the large negative SAM with respect to M2–M3 and TiS–M2 distances are surrounded by the flat region with no SAM and between these regions gentle slopes with small SAM are located. If we assign the region of single-pulse operation we have observed as that of large negative SAM and the CW operation as the flat region, then the region of smaller but still negative SAM which surrounds the region of large negative SAM is considered to correspond to multipulse oscillation. Thus the presence of a proper amount of SAM seems essential to the present multipulse generation. It is also noticed that the magnitude of the SAM without an aperture is considerably smaller than that with an aperture. In this way, the cavity without an aperture seems more favorable to multipulse generation.

On the other hand, the origin of the stepwise change in the pulse interval has not been clarified up to now. Recent simulation implies that the increase in the fourth order dispersion (FOD) causes monotonic increase in pulse separation in the double pulse regime and further increase in FOD results in change into the triple pulse regime [9]. Thus higher order dispersion seems to play an important role. Multipulse generation and stepwise change of the pulse interval will be approached from a different perspective. The stepwise change of the pulse interval is connected essentially with the stabilizing mechanism of multipulse operation. Namely, the fact that the appearance of double pulse oscillation is a gradual function of the pulse separation, means that the oscillating state corresponding to each pulse interval is located on the potential energy surface having a gradual curvature. If we consider the stability of the oscillating state, each oscillating state should form a deep energy hole on the potential energy surface and the transition from one state to another needs large extra energy. If the energy state for each oscillation is continuously distributed, the transition may easily occur and as a result pulse separation is unsettled. Hence, the stability of several hours or more observed in the present study may be thus deeply connected with the stepwise separation change. Such hierarchical structures are characteristic of nonequilibrium systems and more detailed investigation from this viewpoint should be also necessary.

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References