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Abstract. A Yb fiber oscillator producing high-energy femtosecond pulse clusters is reported. Visualized by averaging autocorrelation, the output pulses consist of femtosecond pulse clusters that appear as a picosecond envelope with a ∼100-fs pulse in its center. Using more than 200-m fiber, the pulse energy is scaled up to 450 nJ. This high energy in a cluster of femtosecond pulses enables an important application—laser-induced breakdown spectroscopy. [DOI: 10.1117/1.OE.53.5.051505]

Subject terms: laser-induced breakdown spectroscopy; ultrafast laser applications; plasma spectroscopy; fiber laser.

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In the past two decades, ultrafast fiber laser research has led to the observation of different mode-locking pulse formation regimes, such as soliton,[1] dissipative soliton,[2] and similariton.[3] Efforts to generate high peak power pulses using conventional single-mode fibers (SMFs) are typically limited to 0.5 MW.[4] Under high-gain and high-dispersion conditions, the single-pulses break into multiple pulses, a regime that has been considered as "noise-like" pulses.[5–9] This regime usually generates picosecond or nanosecond long trains of pulses, each containing hundreds of subpulses. Though being avoided by most researchers and considered undesirable, this type of pulse has been applied in metrology, grating based sensing,[10] and supercontinuum generation.[11] Here, we scale up the pulse energy using an unusually long Yb fiber laser cavity. With a more than 200-m-long fiber, the output pulse energy reaches 450 nJ. High pulse energies and the special pulse characteristics make this laser ideal for important applications, such as laser-induced breakdown spectroscopy.

A schematic of the laser design is shown in Fig. 1. This fiber laser is based on an all-normal dispersion cavity using 10/125-μm SMFs. A double clad, 10/125 μm, Yb-doped gain fiber (2 m) is pumped with a diode laser (976 nm) through a fiber combiner. A fiber collimator with 0.35 m passive SMF-II is spliced to the gain fiber to guide laser beam out. Preceding the gain fiber, ∼100- or 200-m-long passive SMF-I is used. A few waveplates and a polarization beam splitter (PBS) are used to enable the nonlinear polarization evolution (NPE)-based mode-locking mechanism.

The laser performance is illustrated in Fig. 2. By adjusting the waveplates and pump power, different mode-locking states can be achieved due to the NPE mechanism. With 100-m-long SMF-I, the repetition rate is 2 MHz [Fig. 2(b)] and the highest output power (640 mW) of stable mode-locking is achieved with 4.5-W pump power, resulting in 320-nJ pulse energy. By lengthening SMF-I to 200 m, the repetition rate is reduced to 1 MHz and the highest pump power for stable mode-locking is 3 W. The corresponding output pulses have average power of 450 mW and 450-nJ pulse energy. The output coupling ratio of the PBS is ∼60%. In both cases, 1 and 2 MHz, the pulse clusters occur as "single pulses." This behavior is very different from when the laser is pumped with higher power and multipulsing occurs preventing one from defining define a repetition rate. The output spectrum is broad and smooth [Fig. 2(a)]. As seen in Fig. 2(b), the pulse train is stable with peak-to-peak fluctuations ∼1%. The mode-locking regime is robust and self-starting.

For output pulses with 640-mW average power at 2 MHz, second harmonic generation (SHG) noncollinear alternating current (AC) measurement is shown in Fig. 2(c). The AC trace, averaging $\sim 10^6$ pulses per delay-time, has a full width at half medium (FWHM) ∼100-fs pulse in the center and a broad picosecond pedestal. This 100 fs is close to the calculated pulse duration based on the output spectrum. Half of the AC trace is shown in Fig. 2(c). The base line drops to below 0.1 at 180 ps. If it was possible to obtain a single-shot AC, one would likely see the output consist of an irregular cluster of femtosecond pulses. The average pulse duration of each subpulse is ∼100 fs and the width of the cluster is as broad as 100 ps.

According to previous study, strong instability can cause the splitting of a single picosecond pulse into many femtosecond subpulses inside the fiber cavity.[7] In our work, the strong instability attributes to the large nonlinearity

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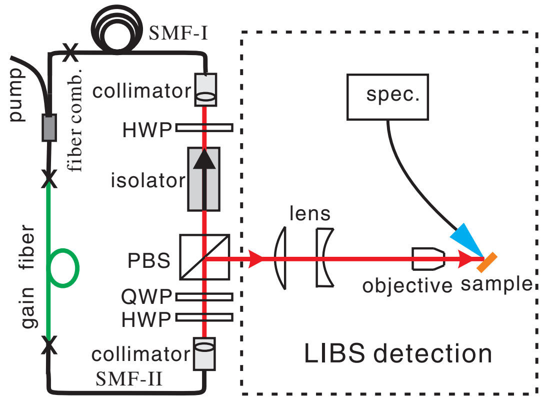

Fig. 1 Experimental setup of the fiber laser (left) and LIBS detection (right). SMF, single-mode fiber; HWP and QWP, half- and quarter-waveplate; PBS, polarization beam splitter.

accumulated through the long fibers and high pulse energy. Each output pulse contains a large number of femtosecond subpulses with different amplitude, phase and pulse duration. The number of subpulses and their time-delay determines the overall time duration of the pulse train. Each pulse train is different, leading to the observed smooth average spectrum and broad AC pedestal.

The phase dependence of the output pulses is also different than that of conventional femtosecond lasers. The integrated SHG signal is found to increase ~5 times by applying second order dispersion ($\sim -60{,}000\ \mathrm{fs}^2$) to the output pulses using a pulse shaper. The ratio between the peak of the spike at the center of the AC and the pedestal within a picosecond can be increased by 2 to 3 times. However, compared to normal femtosecond pulses, the sensitivity of this pulse train to dispersion is more than three orders of magnitude smaller.[5] This implies that these pulses are much less phase sensitive and can be delivered by a fiber with minimal pulse distortion. Group velocity dispersion (GVD) insensitivity enables further energy scaling using even longer intra-cavity fiber. The GVD in the present laser is about $4.6\ \mathrm{ps}^2$.

When the pump power is increased the laser switches from single-cluster output pulses as shown in Fig. 2(b), to random "multipulsing" modes. Therefore, to scale up the pulse energy in this type of laser, a longer cavity is preferable to increasing pump power. Low nonlinearity can be maintained by making sure the long segments of SMF are added before the gain fiber, where the pulse is not yet amplified.

As described above, the laser can produce megahertz repetition rate pulses with high pulse energy. The output picosecond long bursts consist of a number of femtosecond pulses. It has been shown that, for laser-induced breakdown spectroscopy,[12] a pulse train of several nanosecond pulses with either nanosecond or microsecond intervals increases the ablation efficiency compared to that of a single-nanosecond pulse with the same pulse energy.[13,14] It has also been reported that two femtosecond pulses separated by 200 ps lowered the ablation threshold by half compared to a single-femtosecond pulse.[15] In that study, it was observed that two pulses yield a hotter and longer lived plasma, and pulse delays >50 ps were found to yield a greater signal, presumably because they allow time for plasma dissipation.[16]

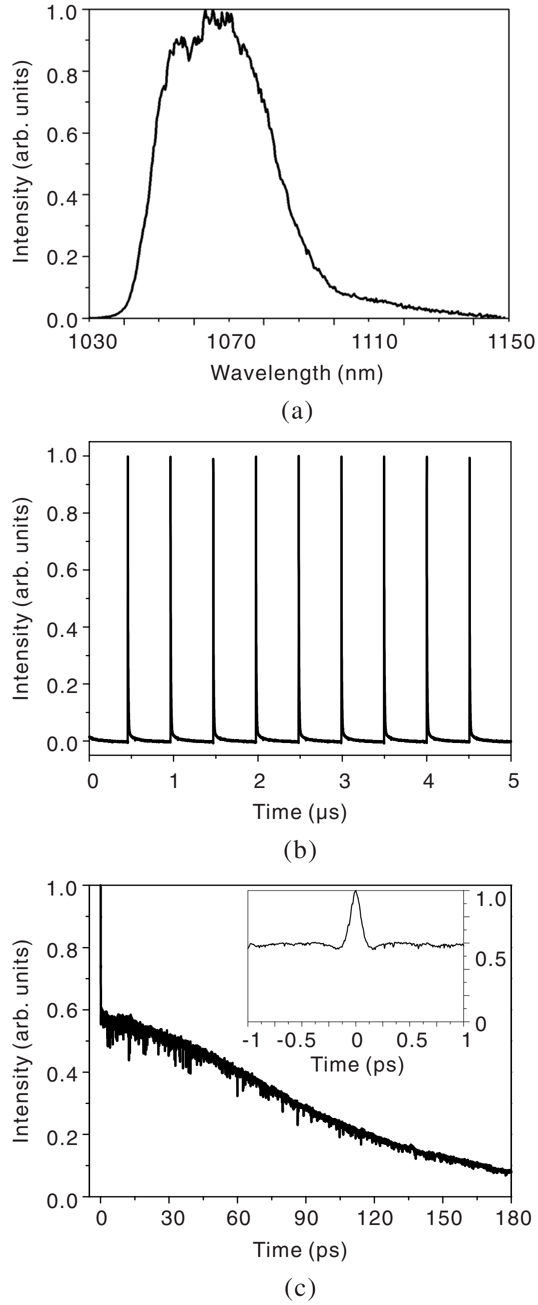

Fig. 2 Laser performance. (a) Output spectrum with average power of 640 mW at repetition rate of 2 MHz; (b) pulse train of laser output at 2 MHz; (c) noncollinear AC trace from 0 to +180 ps. Insect: the same AC trace on a small range from −1 to +1 ps.

From these studies on multiple pulses, and our work on Laser-induced breakdown spectroscopy (LIBS) exploring the effects of spectral bandwidth and pulse shaping,[17] we conclude that our burst laser could be ideal for LIBS, considering its megahertz repetition rate and its bursts containing multiple femtosecond pulses.

As shown in Fig. 1, the output beam of the cavity is collimated with a telescope before being directed to the LIBS detection setup, without pulse compression. A 20× objective (NA = 0.4) is used to focus the beam onto the sample, which is mounted on a spinning wheel (rotation frequency at 133 Hz) to have a fresh spot for ablation. The scattered

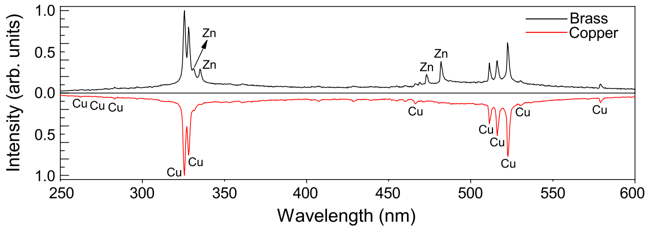

Fig. 3 The LIBS spectra of brass (top) and copper (bottom).

LIBS signal can be directly collected by placing a light collection fiber next to the ablation spot (Fig. 1) and recorded by a compact spectrometer (178 to 876 nm, USB4000, Ocean Optics, Dunedin, Florida).

The results discussed below are obtained using the 2-MHz-cavity output. The Cu and brass pieces are used as samples and their LIBS spectra are compared in Fig. 3. Brass typically contains ∼30% zinc (Zn) and ∼70% Cu. On the brass spectrum, peaks are visible at 330.7, 334.3, 472.3, and 481.1 nm, which are absent in the Cu spectrum. These peaks match Zn emission lines. The other peaks, which are common to both spectra, correspond to Cu emission lines. The spectra shown above are directly recorded without using a gated-spectrometer, and the continuum emission signal is insignificant compared to atomic emission peaks. The low continuum emission also indicates little thermal emission, and it helps limit the heat-affected zone in material processing. The simple detection system requirements further reduce the complexity and cost of the LIBS system. The fast repetition rate at 1 to 2 MHz, comparing to Q-switched lasers, also enhances the accumulation of LIBS signals. Several other samples, such as different aluminum alloys, thin $PbNO_3$ film, and a rock containing lead sulfide (galena), are also tested and they all produced high signal to noise LIBS spectra, which will be demonstrated in another publication.

To further evaluate this system, the dependence of the LIBS signal on pulse fluence is also studied. The measurement shown in Fig. 4 is carried out on Cu. Nonlinear fitting of the experimental data gives a fluence threshold around $0.25\ \mathrm{J/cm^2}$. It is reported that when using a 5-ns Q-switched Nd:YAG laser the threshold fluence for Cu is $1.46\ \mathrm{J/cm^2}$,[18] which is ∼6 times larger than our result. The threshold is also smaller than $0.49\ \mathrm{J/cm^2}$ obtained using a 150-fs Ti:Sapphire chirped-pulse amplifier[19] or $0.5\ \mathrm{J/cm^2}$ obtained using a 35-fs Ti:Sapphire amplifier in our lab.[17]

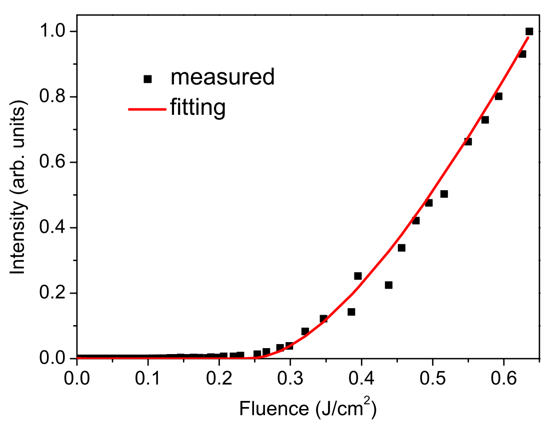

Fig. 4 Threshold measurement. The LIBS intensity for copper as a function of incident laser fluence.

The fluence threshold of ablation is known to increase with $\tau^{1/2}$ for $\tau > 10$ ps,[20–22] where τ is the laser pulse duration. When the pulse duration is less than a few picoseconds, the fluence threshold is near constant for metals.[21,22] Lower threshold implies less pulse energy required for LIBS, which results in a lower background plasma emission. Plasma emission can mask the LIBS signal and eliminating it typically requires a time-gated spectrometer. As discussed above, the threshold measured from our laser is lower than the results of both nanosecond and femtosecond lasers. The observed lower ablation threshold, compared to that of single femtosecond pulses, is probably due to the multiple femtosecond subpulses as indicated by Ref. 15. The lower threshold and high repetition-rate of our laser source results in significant improvements in the efficiency and signal to noise ratio of LIBS spectra. Current portable LIBS systems typically use Q-switched nanosecond fiber lasers.[23] The laser presented here can be developed as an ideal source for a portable LIBS system.

In conclusion, we have demonstrated an Yb fiber oscillator producing up to 450-nJ clusters of femtosecond pulses. Due to the high pulse energy in the pulse cluster and the fact that individual subpulses have pulse durations of ∼100 fs, this laser is capable of generating strong LIBS signals with low fluence threshold and insignificant continuum background. To the best of our knowledge, this is the first time that an Yb fiber oscillator producing noise-like pulses is used for LIBS. One more thing to be noted is that this laser can be easily amplified without prechirp. With a 14-W pump laser at 976 nm applied, the amplified pulse can reach to 3.9 W at 1 MHz, corresponding to 3.9-μJ pulse energy.

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Marcos Dantus has pioneered the use of shaped ultrafast pulses as photonic reagents to probe molecular properties, control chemical reactions and for practical applications such as biomedical imaging, proteomics and standoff detection of explosives. His contributions range from discovery of nonlinear optical properties, invention of laser optimization instruments, and theory to simulate and predict the interaction of molecules lasers. Dantus' development of automated laser pulse compression is enabling research around the world.