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# 1.91 μm Passively continuous-wave mode-locked $Tm:LiLuF_4$ laser ☆

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## 1. Introduction

Tm-, Ho- and Tm,Ho-doped mode-locked bulk lasers at 2 μm are attracted much more attentions due to the emission wavelengths near the absorption peak of water and in the atmospheric window band, which has important applications in surgical operation [1], eye-safe lidar [2], electro-optical countermeasure [3], and remote sensing [4]. In recent years, the ultrafast lasers at 2 μm became also the potential laser sources for chirped-pulse amplification [5], synchronously pumped optical parametric oscillator [6], mid-IR frequency comb [7], supercontinuum generation [5] and material processing [8].

Up to now, the passive mode-locking technology is the main method to obtain ultrafast pulses 2 μm. In 2009, Cho et al. demonstrated a stable and self-starting mode-locked $Tm:KLu(WO_4)_2$ laser using a transmission-type single-walled carbon nanotube (SWCNT) as saturable absorber (SA) [9]. Since then, passive mode locking operations were obtained for a variety of Tm-doped and Tm,Ho-co-doped gain media with different mode-locking devices, such as SESAM, SWCNT, graphene and 2D material (multilayer $MoS_2$, $WS_2$ etc.) and so on. Meanwhile, the novel SAs such as SWCNT [10–11], graphene [12] and 2D material [13] have been explored as passive mode-locking devices. For example, With a graphene saturable absorber, 86-fs pulses was demonstrated from a $Tm:MgWO_4$ laser in 2017 [14]. However, these SAs have relatively high mode-locking threshold because of high loss. With the development of semiconductor technology, the operating wavelength of SESAMs has been extended to infrared range and become one of the most popular devices in 2 μm mode-locked lasers [15] due to its low loss.

As the most widely used mode-locking device, SESAMs were successfully used in Tm-doped and Tm,Ho-co-doped gain media lasers to achieved mode-locking operation. Using tungstate materials as gain media, Lagatsky et al. realized ~100 fs mode-locked operations from $Tm,Ho:NaY(WO_4)_2$ and Tm:KYW oscillators [16–17]. Gluth et al. obtained 3 ps pulses from a Tm:YAG laser [18] and Kong et al. realized a mode-locked $Tm:CaYAlO_4$ laser with 30 ps pulses [19]. Later, Wang et al. demonstrated a $Tm:CaYAlO_4$ oscillator with the pulse duration of 650 fs [20]. In 2017, Luan et al. realized a mode-locked Tm:LuAG laser with the pulse duration of 13.6 ps [21]. Moreover, Tm-doped cubic sesquioxides are also very successful gain materials. In 2012, Lagatsky et al. obtained ~100 fs pulses from $Tm:Sc_2O_3$ and $Tm:Lu_2O_3$ ceramic lasers, respectively [22–23]. In addition, Ma et al. demonstrated a laser diode pumped Tm:CLNGG laser with output of 479 fs pulses [24]. The above mentioned mode-locked lasers have always smooth and non-modulated spectra and the center wavelength is longer than the water absorption peak of 1.95 μm, which can avoid Q-switched instability due to the strong absorption of water in the air [25]. Because of the spectral modulation and the wavelength close to the absorption peak of water, it is hard to achieve continuous-wave mode-locked operation from Tm-doped and Tm,Ho-co-doped fluoride media. Up to now, the only mode-locked operation was obtained from a Tm:GdLiF4 laser [26].

☆ We report, to our knowledge, the first demonstration of continuous-wave mode-locking in a $Tm:LiLuF_4$ laser. By using a GaAs-based semiconductor saturable absorber mirrors (SESAMs), we achieved the mode-locked operation with pulses as short as 14 ps at 1914 nm. The maximum output power is 200 mW. The dependence of the operational parameters on the pump power has been investigated experimentally.

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Compared with other Tm-doped crystal, $Tm:LiLuF_4$ crystal characterized by low phonon energy, big absorption cross section and low absorption for laser, was suitable for low threshold operation [27]. In 2007, Cornacchia et al. investigated the laser performance of $Tm:LiLuF_4$ and obtained the output power of 280 mW with the spectra covering of 1985–2038 nm [28]. In 2009, Xiong et al. achieved the maximum CW power of 7.16 W with the central wavelength of 1.92 μm [29]. Recently, Zou and Zhang et al. demonstrated a Q-switched mode-locked $Tm:LiLuF_4$ laser with $MoS_2$ and Cr:ZnS as saturable absorber, respectively [30–31], however, the continuous-wave mode-locked operation of $Tm:LiLuF_4$ has not been realized. In this paper, to our knowledge, we demonstrated a stable passively continuous-wave mode-locked $Tm:LiLuF_4$ laser for the first time. A stable and self-starting passive continuous mode-locking operation is achieved by using a SESAM from a $Tm:LiLuF_4$ laser with output power of 200 mW and pulse duration of 14 ps at the central wavelength of 1914 nm and a repetition rate of 100 MHz.

## 2. Experimental setup

The experimental setup is shown in Fig. 1. A typical X-shape cavity was used in our experiment.

The pump source is a Ti:sapphire laser with a wavelength range of 740–844 nm, and the output wavelength can be tuned to the 791 nm wavelength by a birefringent filter. L is a focusing lens with a focal length of 120 mm, which is antireflection (AR)-coated at 791 nm. A Brewster-cut 2 at.% doped $Tm:LiLuF_4$ crystal with dimensions of $3 \times 3 \times 12\ mm^3$ is used as the gain medium, which is wrapped with indium foil and tightly mounted on a water-cooled copper heat sink at the temperature of 13 °C to remove the heat deposited inside of crystal. $M_1$ and $M_2$ are plane mirrors, which are highly reflected (>99.9%) at the pump wavelength. $M_3$ and $M_4$ are dichroic mirrors with radii of curvature (ROC) of 100 mm, which have high reflection in the range of 1800–2075 nm (>99.9%) and high transmission in the range of 770–1050 nm (>95%). $M_5$ is a concave mirror with the ROC of 100 mm, which is highly reflected in the range of 1800–2075 nm (>99.9%). $M_6$ is a plane mirror with reflectivity >99.9% at the wavelength range of 1800–2075 nm. OCs are the output couplers, which have transmissions of 1.5%, 3% and 5% in the experiment. A SESAM on GaAs substrate with the non-saturable loss of 2% and the relation time of 10 ps is used to start mode locking. The damage threshold of the SESAM is $4\ mJ/cm^2$. A pair of $CaF_2$ prism ($P_1$ and $P_2$) with a tip-to-tip distance of 300 mm is used for intracavity dispersion compensation.

Based on with ABCD matrix theory, the calculated laser mode size in the crystal is 50 μm, and the radius of the pump spot in the gain medium is set to 24 μm, which is optimized for realization of the efficient fundamental mode output [32]. Meanwhile, the extreme small laser mode is advantageous for laser operation with low threshold.

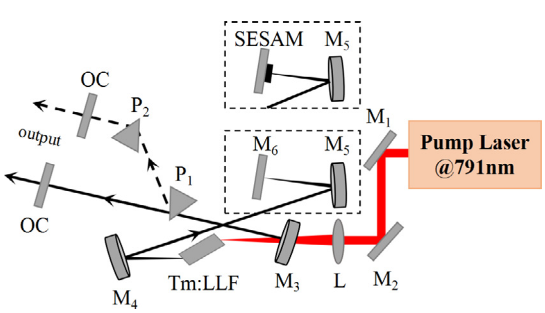


**Fig. 1.** Experimental setup of the passively mode-locked $Tm:LiLuF_4$ laser.

## 3. Analysis and discussion of experimental results

In experiment, the laser threshold is as low as 80 mW (absorbed pump power). To the best of our knowledge, this is the lowest threshold in Tm-doped fluoride lasers. The absorption efficiency in lasing and non-lasing conditions for OCs with different transmissions are measured and compared to investigate the absorption and emission characters of $Tm:LiLiF_4$ crystal, as shown in Fig. 2(a). When the oscillator operated in CW or mode-locking operations, the absorption efficiency is about 65.5%. It shows that the transmission of output couplers had little influence on the absorption efficiency of the gain medium. When the lasing condition is blocked, the absorption efficiency is reduced to 34.3%, which is much lower than the laser running. This is because the stimulated radiation will consume a large number of upper level particles in lasing condition, the absorption rate of crystal is higher. when non laser operation, only spontaneous radiation will consume a few upper level particles, which reduces the efficiency of crystal absorption.

large number of upper level populations is consumed by the laser when it is running. In the state of laser operation, in addition to spontaneous radiation.

The output performance of the $Tm:LiLuF_4$ laser is shown in Fig. 2(b). The output coupler with high reflection is used for the alignment of the cavity. The threshold is only 80 mW (absorbed pump power) when a 1.5% output coupler is used, and the slop efficiency is about 26.8% In this case, the maximum output power of 632 mW is achieved with a optical-to-optical efficiency of 17.1%. When a 3% output coupler is used, the threshold increases to 112 mW and the slop efficiency increases to 33%. The maximum output power is 741 mW, corresponding to an optical-to-optical efficiency of 20%, and when the absorbed pump power is higher than 2.33 W, the laser tends to saturation. With even higher transmission of 5% as the output coupler, 766 mW output is achieved and the optical-to-optical efficiency is 20.8%, and when the absorbed pump power is higher than 2.31 W, the laser tends to saturation. The laser threshold is 131 mW and the slop efficiency is 34.9%.

The mirror of the $M_6$ mirror is replaced by a SESAM to achieve the mode-locking operation. To improve the energy density on the surface of the SESAM, a concave mirror with a radius of 100 mm is used. The stable mode-locking operation is realized by finely adjusting the position of SESAM. The laser mode size on the SESAM is calculated to be about 140 μm, corresponding to the energy fluence about $336\ \mu J/cm^2$. It is much larger than the saturable fluence ($70\ \mu J/cm^2$) of SESAM. A pair of $CaF_2$ prisms is inserted into the cavity to compensate the intracavity dispersion and shorten the pulse duration. With a 1.5% output coupler, the cavity begins output with Q-switched pulse trains when the absorbed pump power is 228 mW. Further increase the absorbed pump power to 822 mW, the Q-switched mode-locking operation is achieved. The stable mode-locking operation is realized at the absorbed pump power of 1.9 W. The maximum output power of 200 mW is achieved at 2.4 W absorbed pump power, corresponding to an optical-to-optical efficiency of 5.4%.

The mode-locked pulse train is measured with a high-speed photodiode (PD) and a 200-MHz digital oscilloscope (RIGOL, DS4024), as shown in Fig. 3. It shows the mode-locked pulse train has a repetition rate of 100 MHz, corresponding to a maximum pulse energy of 2 nJ.

The spectra of the mode-locked pulses are measured with a commercial spectrometer (AvaSpec-NIR256-2.5TEC). The full