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# Passively Q-Switched Mode-Locked Tm,Ho:$CaYAlO_4$ Laser Based on Double-Walled Carbon Nanotube Saturable Absorber

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A passively Q-switched mode-locked (QML) operation in Tm,Ho:$CaYAlO_4$ bulk laser was demonstrated experimentally by employing double-walled carbon nanotubes (DWCNTs) as a saturable absorber. The laser is pumped by a self-made wavelength tunable Ti:sapphire laser, and the pump threshold of Tm,Ho:$CaYAlO_4$ laser was measured at 677 mW using transmittance of 1.5% output coupler. A stable QML operation state was achieved when the absorption pumping power reached 1,958 mW. When the pumping power reached 2.6 W, the maximum output power was 64 mw with a central wavelength of 2,085 nm, the corresponding repetition frequency of mode-locked pulse was 98.04 MHz, and the modulation depth in Q-switching envelopes is close to 100%.

**Keywords: Tm,Ho:$CaYAlO_4$ laser, passively Q-switched mode-locked, double-walled carbon nanotube, saturable absorber, resonant cavity**

**OCIS codes: 140.3580 (Lasers, solid-state), 140.3070 (Infrared and far-infrared lasers), 140.3380 (Laser materials), 140.4050 (Mode-locked lasers), 140.3410 (Laser resonators)**

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## INTRODUCTION

Recently, thanks to the rapid development of laser technology, a variety of lasers, including medium infrared ultrafast lasers [1, 2], ultrafast fiber lasers [3, 4] are playing an important role in more and more fields [5, 6]. Among them, ultrafast solid-state lasers emitting around eye-safe 2 μm exhibits the potential applications in LIDAR, biomedicine, time-resolved spectroscopy, atmospheric remote sensing, nonlinear frequency conversion, optical communications, etc. [7–9]. Crystal, ceramic, and glass materials doped with thulium ($Tm^{3+}$), holmium ($Ho^{3+}$) ions [10], or $Tm^{3+}$, $Ho^{3+}$ co-doped are currently the most promising candidates for 2-μm mid-infrared laser sources. The passive mode-locking technique with saturable absorber (SA) [11–13] is the most widely convenient and low-cost method to obtain ultrafast lasers at 2-μm wavelength. Up to now, SAs such as semiconductor saturable absorber mirrors (SESAMs) [14–16], carbon nanotubes [17–19], graphene [20–22], and transition metal dichalcogenides (TMDs) [23, 24] have been adopted for passive Q-switching or mode-locking operations. However, SESAMs have the disadvantage of having narrow bandwidth, complex fabrication, and high cost to limit the application and development of mid-infrared ultrafast lasers. Therefore, it is very important to develop 2-μm wavelength ultrafast lasers based on new materials.

In recent years, a batch of new 2D nanomaterials with unique properties has received widespread attention. Among them, carbon nanotubes are favored as new SA in the field of ultrashort pulse laser due to their excellent electrical, optical, and mechanical properties. According to the number

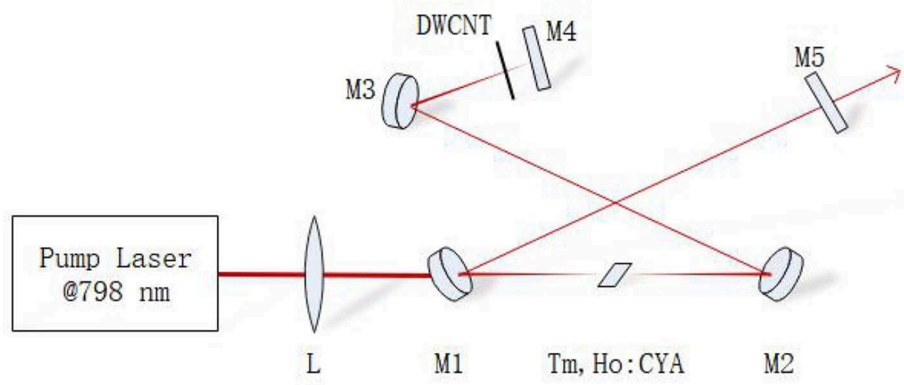


**FIGURE 1 |** The experimental setup of Tm,Ho:CaYAlO$_4$ passively QML laser structure.

of graphite layers, carbon nanotubes are divided into single-walled carbon nanotubes (SWCNT) and multiwalled carbon nanotubes [25]. It has been reported that SWCNT-SAs can passively mode lock in 0.8–2-μm laser [26–28]. As the simplest multiwalled carbon nanotubes, double-walled carbon nanotubes (DWCNTs) are made of two layers of graphite, which are coiled according to a certain helix angle. The diameters of the inner and outer walls are 0.8–1.1 and 1.6–1.8 nm, respectively, and the spacing between the inner and outer layers is 0.34–0.39 nm [29]. Under the same irradiation condition, DWCNT conductivity is better due to its higher chemical stability and smaller energy gap than SWCNT [30]. Meanwhile, DWCNT has the characteristics of relatively low cost, relatively short relaxation time, and the advantages for mass production. Therefore, DWCNT-SAs has ultra-short recovery time, wider absorption band, and higher damage threshold in a 1–2-μm band [31]. Now, the reports on mode-locked DWCNT-SAs are mainly focused on the solid-state and fiber lasers at 1-μm band [32, 33], while the reports on mode-locked DWCNT-SAs at 2-μm band are few, and the output power is lower than 200 mW [34–36]. For example, Wang et al. [37] achieved a mode-locked operation of 0.98 ps in $Tm^{3+}$-doped silica fiber, and Qu et al. [38] achieved a mode-locked operation in Tm:YAP laser. Over the years, our group has been devoted to the research of ultrafast laser technology in the mid-infrared band. In 2018, our group realized the Q-switched and mode-locked simultaneous operation with low threshold based on the DWCNTs in the Tm,Ho:LLF laser, and the maximum output power was 234 mW [39].

CaYAlO$_4$(CYA) crystal belongs to the perovskite structure, which is an excellent laser medium matrix material fabricated by the Czochralski method [40]. Tm,Ho:CaYAlO$_4$ crystals have higher absorption efficiency and wider tuning width, and their main absorption peaks are 691, 797, 1,212, and 1,694 nm [41]. Currently, there are few researches on related mode locking of this crystal. In 2018, only Zhao et al. realized continuous mode-locking operation in Tm,Ho:CaYAlO$_4$ laser based on SESAM [10].

In this paper, a stable simultaneously Q-switched mode-locking (QML) operation was experimentally demonstrated for the first time in Tm,Ho:CaYAlO$_4$ crystal by using DWCNTs-SA. The pump source was a self-made wavelength-tunable Ti:sapphire solid laser. With 1.5% output coupler, the maximum output power of QML is 64 mw at the central wavelength of 2,085 nm, the repetition frequency of mode-locked pulse in Q-switched envelope is 98.04 MHz, and the modulation depth was close to 100%.

## EXPERIMENTAL SYSTEM

The experimental setup of the passive mode-locking Tm,Ho:CaYAlO$_4$ laser is shown in **Figure 1** [9]. A typical X-type five-mirror cavity structure is adopted to obtain better pattern matching effect, which is composed of a typical X-type four-mirror folded cavity and focused concave mirror. The laser is pumped by a self-made Ti:sapphire solid-state laser with an output wavelength of 798 nm. The laser crystal of Tm,Ho:CaYAlO$_4$ with 6% $Tm^{3+}$ and 0.5% $Ho^{3+}$-doped was cut at the angle of Brewster. Its size is 3 × 3 × 4 mm, and the strongest absorption peak is 798 nm. In order to reduce the thermal lensing effect of crystal and mitigate the thermal load, it is necessary to cool the laser crystal to ensure the stable operation of the laser. Here, the laser crystal is wrapped in indium foil and mounted in a copper heat sink, which is cooled by circulating water at a constant temperature of 12°C [42]. The standard X-folded cavity consisted of M1, M2, M3, M4, and an output coupler (M5). In **Figure 1**, M1 and M2 are 2-μm pump mirrors produced by Layertec company with curvature radii of 100 and 75 mm, respectively, whose transmittance are higher than 95% in the wavelength range from 770 to 1,050 nm, and reflectivity is higher than 99.9% at 2-μm wave bands. M3 is a plane-concave reflector with the curvature radius of concave surface of 100 mm, and M4 is a planar reflector. The reflectivity of both flat concave mirror M3 and flat reflector M4 is >99.9% for oscillating light at 2-μm wave bands. M5 is an output coupler with partial transmission for oscillating light. In the experiment, the output mirror with a transmittance of 1.5% was selected to obtain a high intracavity power density. DWCNTs were inserted before the M4 plane mirror as SA. The collimated pump light is incident into the Tm,Ho:CaYAlO$_4$ crystal by a focusing lens ($f$ = 150 mm) with higher than 95% transmittance for 798 nm. The laser beam diameter is 54 μm on the surface of SA, which

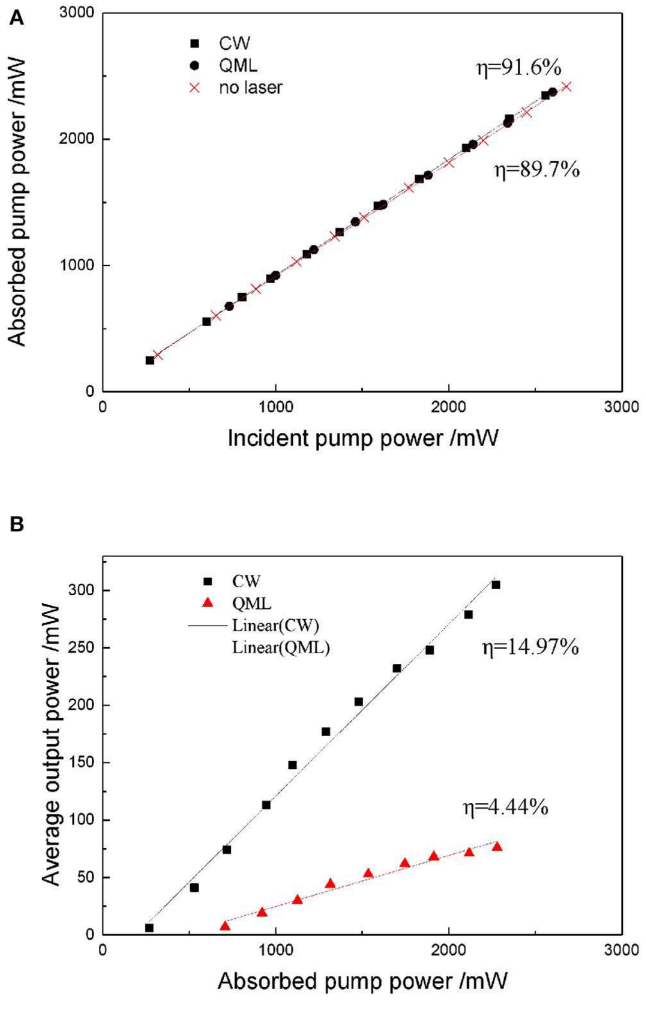


**FIGURE 2 |** Experimental data diagram. **(A)** The dependence of absorbed power on incident pump power. **(B)** The dependence of average output power on absorbed pump power under CW and QML.

is calculated using the laser cavity mode ABCD propagation matrix theory.

## EXPERIMENTAL RESULTS AND DISCUSSION

The absorption and output characteristics of the laser are shown in **Figure 2**. First, the absorption efficiency of the laser crystal of Tm,Ho:$CaYAlO_4$ is shown in **Figure 2A** [43]. It can be seen that the absorption efficiency of laser crystal to the pump light at 798 nm is 89.7% due to a large amount of pump light that is absorbed by the crystal when there is no laser running in the cavity. When continuous wave (CW) operation in the cavity is realized, the absorption efficiency of the laser crystal increases to about 91.6% due to the large number of upper-level particles returning to the lower level under stimulated radiation. After inserting the DWCNT-SA into the cavity, the operating state of QML is achieved, and the absorption efficiency of the laser crystal did not change significantly and still remained around 91.6%.

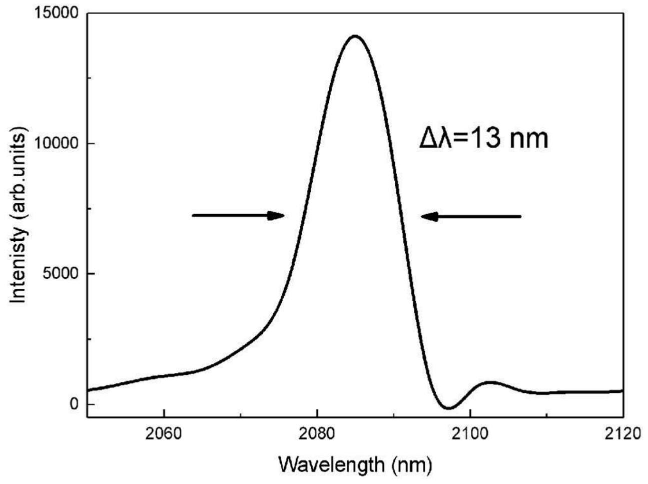


**FIGURE 3 |** The emission spectrum of QML Tm,Ho: $CaYAlO_4$ laser.

Next, the average output power as a function of the absorbed pump power under CW and QML is plotted in **Figure 2B**. In the experiment, 1.5% output coupling mirror is used. When the laser is in CW operation, the laser threshold power is 249 mW, the maximum output power is 301 mW, and the corresponding slope efficiency is 14.97%. When DWCNT-SA was inserted before M4 in the cavity, the laser threshold power increased to 677 mW. When the absorbed pump power reached 1,958 mW, laser entered a stable QML operation, and the maximum output power under the same conditions was 64 mW, which has a corresponding slope efficiency of 4.44%. Here, the main reason why we choose 1.5% output coupler is that it can provide high intracavity power to start QML.

**Figure 3** shows the typical spectra of the mode-locked pulse measured with a spectrometer (AvaSpecNIR256-2.5TEC) [9]. As can be seen from **Figure 3**, the central wavelength of a mode-locked laser pulse is 2,085 nm, and the full width at half-maximum bandwidth is about 13 nm. A 2-μm fast photodiode (ET-5000) was used to connect a 200-MHz digital oscilloscope (RIGOL, DS4024) to detect QML pulse sequences. **Figure 4** shows the QML pulse sequences, which is obtained by scanning times of (a) 1 ms, (b) 100 μs, and (c) 10 ns. When the output power reaches the maximum, the pulse width of the Q-switched envelope is 12 μs, the repetition frequency is 83.33 kHz, the frequency of the mode-locked pulse is 98.04 MHz, and the modulation depth of the mode-locked pulse is close to 100%, which is consistent with the theoretical repetition frequency corresponding to the 1.5-m cavity length.

QML is in the transition state from Q-switch to CW mode locking [44–46], Since an autocorrelator is only suitable for measuring the pulse width of a CW mode-locked pulse, a QML pulse cannot obtain autocorrelation envelope because of the envelope modulation of kilohertz, so we can only estimate the pulse duration roughly in theory.

Hence, formula (1) is used to calculate the width of the mode-locked pulse [47].

$$t_m = \sqrt{{t_r}^2 + {t_p}^2 + {t_o}^2} \tag{1}$$

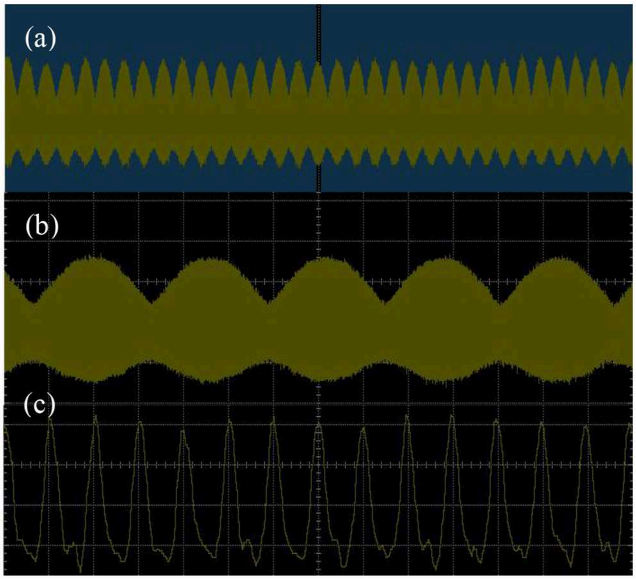


**FIGURE 4 |** Mode-locked pulse sequence recorded in **(A)** 1 ms, **(B)** 100 μs, and **(C)** 10 ns per division (div) timescales.

Here, $t_m$ is the rising edge time of the measured mode-locked pulse, $t_r$ is the rising edge time of the actual mode-locked pulse, $t_p$ is the rising edge time of the photodetector, and $t_0$ is the rising edge time of the oscilloscope. In the experiment, the rising edge time of the mode-locked pulse is about 2.1 ns, and the rising edge time of the photodetector is about 35 ps. $t_0$ can be estimated as 2,000 ps using formula (2) as follows.

$$t_0 \times W_b(200MHz) = 0.35 \sim 0.4 \quad (2)$$

Among them, $W_b$ is the bandwidth of the oscilloscope, which is 200 MHz in the experiment. Therefore, it can be calculated that the rise time of a mode-locked pulse is about 639.35 ps. Since the actual mode-locked pulse width is about 1.25 times the rising edge time, it is calculated as 799.2 ps.

## CONCLUSION

A passively Q-switched mode locking operation is realized experimentally by using DWCNT-SA in Tm,Ho:CaYAlO$_4$ all-solid laser for the first time. After DWCNT-SA was added into the resonator cavity, the pump threshold of Tm,Ho:CaYAlO$_4$ solid-state laser was measured as 677 mW using the transmittance of 1.5% output coupler. When the absorption pumping power reached 1,958 mW, Tm,Ho:CaYAlO$_4$ solid state laser entered a stable QML operation state. When the pump power was 2.6 W, the maximum output power was 64 mw at the central wavelength of 2,085 nm, the mode-locked pulse repetition frequency is 98.04 MHz, and the modulation depth is close to 100%. The experimental results show that DWCNT-SA can be used as a quick starting element for passively QML solid state laser of 2-μm band, which has an important development and application value.

## DATA AVAILABILITY STATEMENT

All datasets generated for this study are included in the article/supplementary material.

## AUTHOR CONTRIBUTIONS

YZ was the author of the experimental scheme and the general director of the project. WL was the specific guidance for postgraduates during the experiment. DQ, who is a doctoral student, and RS and CC, who are graduate students of the research group, have implemented the experimental scheme.

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**Conflict of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.