

# Passive mode locking of 2.09 $\mu\text{m}$ Cr,Tm,Ho:Y<sub>3</sub>Sc<sub>2</sub>Al<sub>3</sub>O<sub>12</sub> laser using PbS quantum-dot-doped glass

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Passive *Q*-switched mode locking of a 2.09  $\mu\text{m}$  flashlamp-pumped Cr<sup>3+</sup>, Tm<sup>3+</sup>, Ho<sup>3+</sup>:Y<sub>3</sub>Sc<sub>2</sub>Al<sub>3</sub>O<sub>12</sub> laser by use of a phosphate glass doped with PbS quantum dots of 5 nm in radius was demonstrated. Mode-locked pulses of 290 ps in duration and up to 0.5 mJ in energy were registered. © 2009 Optical Society of America  
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Lasers emitting pulse radiation at wavelengths around 2  $\mu\text{m}$  are important for various applications in medicine, chemical pollution analysis, meteorology, etc. Use of saturable absorbers (SAs) for pulse generation allows such lasers to be more reliable and compact. There are several materials demonstrating absorption saturation (bleaching effect) in this spectral range [1–4]. However, using SAs based on these materials, only passive *Q* switching of holmium lasers was demonstrated [1–4]. Recently, passive mode locking of Ti:sapphire laser-pumped 2  $\mu\text{m}$  Tm,Ho:Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> and Tm,Ho:KY(WO<sub>4</sub>)<sub>2</sub> lasers using semiconductor SA mirrors based on the saturable absorption of intersubband [5] and interband [6] transitions in quantum wells has been reported. Ultrashort pulses of 3.3 ps in duration were registered in the Tm,Ho:KY(WO<sub>4</sub>)<sub>2</sub> laser with InGaAsSb quantum wells as the absorber [6]. In this Letter, we report a glass doped with lead sulfide (PbS) quantum dots (QDs) as another type of material for the passive mode locking of holmium lasers. Using such glasses, passive mode locking of the various solid-state lasers emitting at the wavelengths from 1.03 to 1.55  $\mu\text{m}$  was demonstrated [7–12]. The absorption saturation at the wavelengths through the first excitonic absorption band of PbS QDs was utilized. Owing to the quantum confinement effect, it is possible to shift the spectral position of the PbS QDs' first excitonic absorption band to the  $\sim 2$   $\mu\text{m}$  range by varying the size of QDs. Glasses containing PbS QDs of appropriate size were successfully used for the passive *Q* switching of Ho:Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> and Ho:Y<sub>3</sub>Sc<sub>2</sub>Al<sub>3</sub>O<sub>12</sub> lasers [4]. Here, we present the passive *Q*-switched mode locking of the flash-lamp pumped 2.09  $\mu\text{m}$  Cr<sup>3+</sup>, Tm<sup>3+</sup>, Ho<sup>3+</sup>:Y<sub>3</sub>Sc<sub>2</sub>Al<sub>3</sub>O<sub>12</sub> (Cr,Tm,Ho:YSAG) laser by use of the glass doped with PbS QDs.

A glass sample was prepared using a P<sub>2</sub>O<sub>5</sub>–Na<sub>2</sub>O–ZnO–AlF<sub>3</sub>–Ga<sub>2</sub>O<sub>3</sub> system by the glass batch-melting technique. The conditions of a secondary (after glass annealing) heat treatment were chosen to obtain the PbS QDs with the absorption peak corresponding to the first excitonic resonance located

in the region of 2  $\mu\text{m}$ . The absorption spectrum of the glass with PbS QDs is presented in Fig. 1(a). The dependence of the absorption band maximum spectral position on the PbS QDs size from [13] allows estimation of their mean radius as 5.0 nm.

The absorption saturation measurement of the PbS QDs doped glass at the wavelength of 2.1  $\mu\text{m}$  was performed using a passively *Q*-switched Ho:Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> laser. The laser worked in a TEM<sub>00</sub> mode and emitted pulses with energy of 3 mJ and duration of 55 ns.

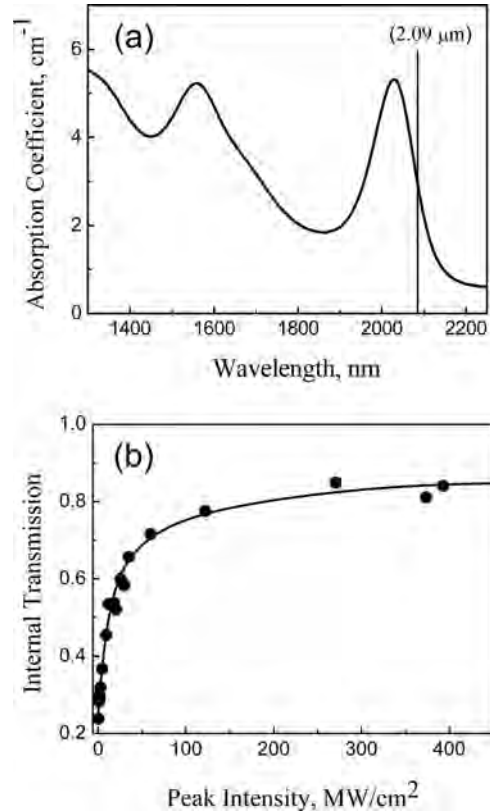


Fig. 1. (a) Absorption spectrum and (b) intensity-dependent transmission at the wavelength of 2.1  $\mu\text{m}$  of the PbS quantum-dots-doped glass. A vertical bar in (a) shows the Cr,Tm,Ho:YSAG laser emission wavelength.

The input intensity  $I$  in the glass sample was varied in the range of  $I=0.1\text{--}210\text{ MW/cm}^2$  by translating the sample along the optical axis of the experimental setup ( $Z$ -scan-like technique). Figure 1(b) presents results of the intensity-dependent transmission measurements of the studied PbS QDs doped glass. A considerable increase of the sample transmission at a high pump level (bleaching effect) is observed. The optical damage of the sample was not detected at the maximum input intensity. Since the pump pulse duration (55 ns) was appreciably longer than the PbS QDs' bleaching relaxation time [7–11], the experimental data were analyzed in the framework of the fast four-level SA model, taking into account the excited-state absorption effect [14]:

$$\ln \frac{T}{T_0} - \left( \frac{\sigma_a}{\sigma_{\text{esa}}} - 1 \right) \ln \left[ \left( 1 + \frac{\sigma_{\text{esa}} I}{\sigma_a I_s} \right) / \left( 1 + \frac{\sigma_{\text{esa}} I}{\sigma_a I_s} T \right) \right] = 0, \quad (1)$$

where  $T_0$  and  $T$  are the small-signal and the intensity-dependent transmissions of the PbS QDs doped glass, respectively;  $\sigma_a$  and  $\sigma_{\text{esa}}$  are the ground-state and excited-state absorption cross-sections, respectively;  $I_s$  is the sample absorption saturation intensity. All calculations were carried out for the Gaussian temporal and spatial shape of laser pulses.

The best fit of the numerical calculation results to the experimental data was achieved with the values of  $I_s=2\pm 0.2\text{ MW/cm}^2$  and  $\sigma_{\text{esa}}/\sigma_a=0.08\pm 0.01$ . These data are in agreement with the data reported for the PbS QDs of smaller sizes [7–12,15].

A 0.5-mm-thick SA with the internal small-signal transmission of  $T_0=75\%$  and without antireflection coatings have been prepared from the studied PbS QDs doped phosphate glass. The 102-cm-long laser cavity has been composed from a curved high reflector (radius of curvature,  $r=100\text{ cm}$ ) and a flat output coupler with reflectivity of 70% at the lasing wavelength. Such cavity is close to the semispherical configuration, which is preferable to obtain the TEM<sub>00</sub> mode laser operation [16]. Additionally, the intracavity mode radius is varied in the wide range, which allows one to change the ratio of mode areas in the gain medium and the SA,  $\rho_{\text{ga}}$  and is important to achieve a laser mode-locking operation [17]. A flashlamp-pumped water-cooled Cr,Tm,Ho:YSAG rod of  $\varnothing 4 \times 73\text{ mm}^2$  in size with the antireflection coatings has been used as the gain medium. The laser operated at the 0.5 Hz repetition rate. The output radiation has been converted into the second harmonic in a LiNbO<sub>3</sub> nonlinear crystal and detected by a Ge avalanche photodiode and a 500 MHz Tektronix 3052TDS digital oscilloscope. Time resolution of the registration system was  $\sim 1\text{ ns}$ .

$Q$ -switched mode locking of the Cr,Tm,Ho:YSAG laser was achieved with the SA placed between the laser rod and the output coupler perpendicular to the cavity optical axis. The typical mode-locked pulse train of 6 mJ total energy is presented in Fig. 2(a).

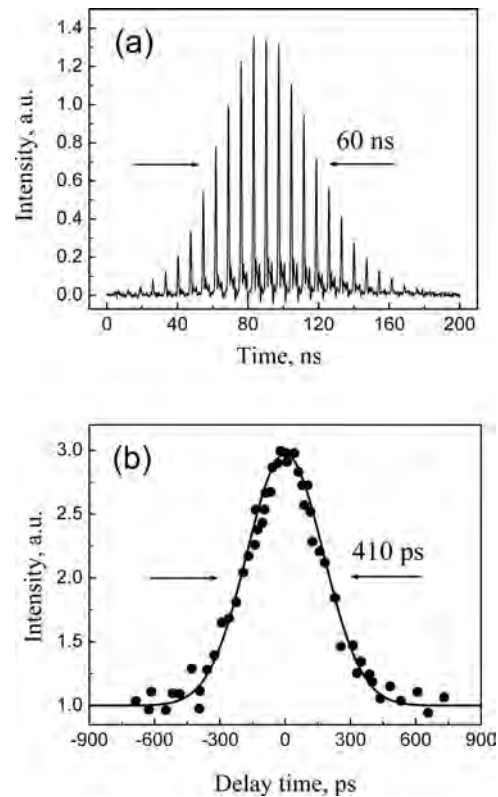


Fig. 2. (a) Train of mode-locked pulses from the Cr,Tm,Ho:YSAG laser with the PbS quantum-dots-based saturable absorber and (b) the intensity autocorrelation function. Symbols in (b) are the experimental data; the curve is the fit assuming the Gaussian temporal profile of a single ultrashort laser output pulse.

The train has a  $Q$ -switched pulse envelope with a 100% modulation depth. Ultrashort pulses are separated by the 7.2 ns cavity round-trip time. The  $Q$ -switched envelope has a FWHM of 60 ns. The average single pulse energy of 0.3 mJ (and the maximal one of 0.5 mJ for a central pulse in the train) has been obtained. No optical destruction or photodarkening of the SA during long-term operation was detected. Stable laser  $Q$ -switched mode-locking operation was observed for the focusing parameter  $\rho_{\text{ga}}=1.6\text{--}2.5$  ( $Q$ -switched pulse with 100% modulation depth). Higher increase of the intracavity radiation focusing in the SA  $\rho_{\text{ga}}$  led to the instability of the  $Q$ -switched mode-locking operation. Namely, the modulation depth of the  $Q$ -switched pulse tended to minimize followed by the laser switching to the  $Q$ -switching regime without mode locking. Such laser behavior is in agreement with the flashlamp-pumped lasers passive mode-locking theory [17]. The latter predicts that the mode locking may be achieved only simultaneously with the  $Q$  switching ( $Q$ -switched mode locking) and that the most efficient and stable mode-locking operation may be obtained when the laser operates in the regime of a small exceeding of the so-called second threshold (the threshold of the mode-locking operation) [17]. An increase of  $\rho_{\text{ga}}$  leads to an enhancement of the exceeding of the second threshold and, consequently, to the appearance of instabilities in the laser-operation regime [17]. The

condition of the second threshold was not satisfied for  $\rho_{\text{ga}} < 1$  when the SA was placed between the gain medium and the high reflector, and the laser worked in free-running mode.

The duration of a single ultrashort pulse was measured using the autocorrelation function measurement method. The autocorrelation function has been registered by an Si photodiode based on the nonlinear effect of the two-photon absorption of the 2.09  $\mu\text{m}$  laser radiation in Si [18]. The measured autocorrelation function is presented in Fig. 2(b). The FWHM of the function is 410 ps. Assuming the Gaussian temporal profile of a single mode-locked laser pulse, its average duration was estimated as  $\Delta t_p = 410/\sqrt{2} \approx 290$  ps.

The estimation of the SA modulation depth can be made. Maximum single mode-locking pulse energy of 0.5 mJ corresponds to the 2.07  $\text{GW}/\text{cm}^2$  of the intracavity input intensity on the SA. According to Eq. (1), under such excitation the SA with the  $T_0=75\%$ ,  $I_s=2 \text{ MW}/\text{cm}^2$ , and  $\sigma_{\text{esa}}/\sigma_a=0.08$  is bleached to the value of the internal transmission  $T=97\%$ , which corresponds to the SA modulation depth of 22% and the internal nonsaturable losses of 3%.

It was shown that the temporal characteristics of the bleaching relaxation of PbS QDs depend on the QDs' mean size and the spectral position of the laser wavelength relative to the maximum of the first excitonic absorption band (e.g., see [11,19]). That is why we believe that the SA based on the glass doped with PbS QDs of the appropriate size will allow generation of the shorter mode-locked pulses from the holmium lasers. Improvement of the laser output characteristics may be also achieved by the use of antireflection coatings on the SA and by the proper choice of the SA initial transmission and the optimization of the output coupler reflectivity. Furthermore, the cw mode locking of different solid-state lasers (diode- and laser-pumped) using glasses doped with PbS QDs have been reported [8,10,12]. Therefore, the realization of a cw mode-locked 2  $\mu\text{m}$  laser with the PbS QDs SA may be awaited.

In conclusion, we have achieved the passive Q-switched mode locking of the flash-lamp pumped  $\text{Cr}^{3+}, \text{Tm}^{3+}, \text{Ho}^{3+}:\text{Y}_3\text{Sc}_2\text{Al}_3\text{O}_{12}$  laser using the glass doped with lead sulfide quantum dots of 5 nm in the mean radius. Mode-locked pulses of 290 ps in duration and up to 0.5 mJ in energy have been obtained at the wavelength of 2.09  $\mu\text{m}$ . The SA based on the PbS QDs doped glass is characterized with the values of the absorption saturation intensity  $I_s=2 \text{ MW}/\text{cm}^2$  and the ratio of the excited-state to ground-state ab-

sorption cross sections  $\sigma_{\text{esa}}/\sigma_a=0.08$  at the wavelength of 2.1  $\mu\text{m}$ .

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