



# Study on the dopant concentration ratio in thulium-holmium doped silica fibers for lasing at 2.1 $\mu\text{m}$

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**Abstract:** We present the fabrication and laser performance of thulium-holmium co-doped silica fibers when cladding pumped at  $\sim 790$  nm. By using the hybrid gas phase-solution doping process in conjunction with the MCVD preform fabrication technique, the doping concentration and the Tm:Ho ratio were varied to study the energy transfer efficiency from  $\text{Tm}^{3+}$  to  $\text{Ho}^{3+}$ . Our study indicates that for a thulium concentration that has resulted in an efficient two-for-one cross-relaxation process with 790 nm pumping, and while maintaining a Tm:Ho concentration ratio in the range  $\sim 10$  to 20, the energy transfer efficiency has reached above 75%. In a free-running laser cavity, the pump power limited laser output of 38W with a slope efficiency of 56% at 2.1 microns is demonstrated.

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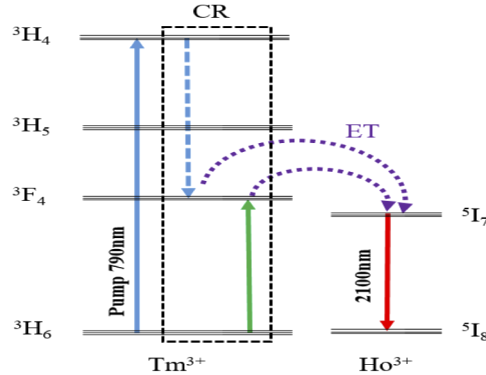
## 1. Introduction

Over the last decade, thulium-doped fiber lasers (TDFLs) and holmium-doped fiber lasers (HDFLs) have shown significant progress in the longer side of the near infra-red region (from 1800 nm to 2200 nm) for applications in different fields especially in security, medicine, materials processing, and LIDAR systems for remote sensing [1–5]. In TDFLs, when cladding pumped at 79x nm, where the high power diodes are available, an excited  $\text{Tm}^{3+}$  ion in the  $^3\text{H}_4$  manifold interacts with a nearby ground state ion in the  $^3\text{H}_6$  manifold, producing two excited ions to lase from the  $^3\text{F}_4$  manifold (two-for-one cross-relaxation process), reaching a quantum efficiency up to 200% in the two micron region [6].

In contrast, the  $\text{Ho}^{3+}$  ions have no absorption bands where high power diodes are currently available [7]. For accessing wavelengths beyond 2.1  $\mu\text{m}$ , HDFLs are a preferred choice. Lasers operating in a window of high atmospheric transmission between 2.1–2.5  $\mu\text{m}$  open up opportunities for free-space communications and range-finding. One of the absorption bands for HDFLs lies at  $\sim 1.95$   $\mu\text{m}$  ( $^5\text{I}_7$  manifold). Consequently, an in-band pumping scheme using TDFLs as the pump source is commonly used [8,9]. However, the laser efficiency of 790 nm diode pumped TDFLs operating at  $\sim 1.95$   $\mu\text{m}$  is somewhat less than when operating beyond 2  $\mu\text{m}$  [10,11], and as a result of that the overall electrical-to-optical conversion efficiency of lasers operating at 2.1  $\mu\text{m}$  could suffer. Moreover, the implementation of this pumping scheme brings additional complexity to fiber fabrication due to the need for an all-glass fiber structure with a fluorine doped cladding for low-loss pump guidance, as low-index polymer used in standard double clad fibers will incur a strong absorption in the 2  $\mu\text{m}$  wavelength region [12–15].

Alternative to the in-band pumping scheme, co-doping of silica fibers with thulium and holmium can be considered [16,17]. In that case, the 79x nm pump can be utilized to excite thulium ions and promote the two-for-one cross-relaxation process, followed by a dominant donor-acceptor energy transfer mechanism from thulium  $^3\text{F}_4$  manifold to holmium  $^5\text{I}_7$  manifold

[18,19] as shown in Fig. 1. Laser sources based on the Tm:Ho co-doped silica fiber have been reported with a slope efficiency up to 42% [20]. Also, it has been suggested that an improvement in the laser performance could be achieved with an optimized fiber core composition.



**Fig. 1.** Energy levels diagram of a Tm:Ho co-doped system.

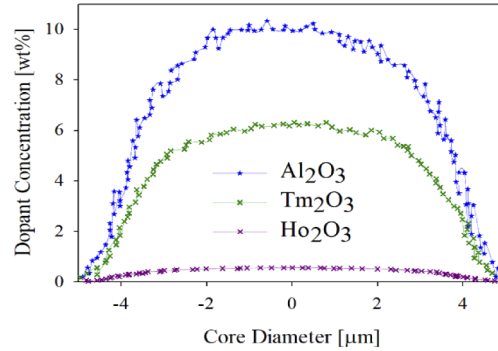
In this work, we studied the effect of thulium and holmium dopant concentrations on the energy transfer efficiency between donor ( $\text{Tm}^{3+}$ ) and acceptor ( $\text{Ho}^{3+}$ ) ions and the laser performance at 2.1  $\mu\text{m}$  in Tm-Ho co-doped silica fibers when cladding pumped at 79x nm. In our previous reports, we have shown that thulium-doped fibers (TDFs) fabricated by a hybrid gas phase-solution doping technique in combination with the MCVD process, allow a characteristic flat top refractive index profile, promoting higher laser efficiencies at  $\sim 2 \mu\text{m}$  [11]. Tm-Ho co-doped silica fibers presented here were fabricated by the hybrid gas phase-solution doping process. Our results indicate that for a  $\text{Tm}^{3+}$  concentration of  $\sim 5\text{wt}\%$  and while maintaining a Tm/Ho concentration ratio of  $\sim 10$ , an efficient two-for-one cross-relaxation process in thulium ions as well as a good energy transfer efficiency ( $> 75\%$ ) from  $\text{Tm}^{3+}$  to  $\text{Ho}^{3+}$  ions can be reached, resulting in laser efficiencies above 56% operating beyond 2100 nm.

## 2. Experimental and results

Preforms with 2 wt% and 5 wt% of  $\text{Tm}^{3+}$  concentrations and three different Tm/Ho concentration ratio of 5, 10 and 20, respectively, were fabricated by the MCVD process in combination with a hybrid gas phase-solution doping technique. In addition, holmium free Tm-doped preforms with the same  $\text{Tm}^{3+}$  concentrations as above were presented. It is worth mentioning that a  $\text{Tm}^{3+}$  concentration greater than 2 wt% is necessary to utilize two-for-one cross-relaxation process in TDFL with 790 nm pumping. In the first step, a cladding of silicon oxide ( $\text{SiO}_2$ ) was deposited inside a substrate tube (Suprasil F-300) followed by an aluminosilicate soot layer deposited through the vapor phase technique. The aluminum chloride ( $\text{AlCl}_3$ , 99.999%) was heated at  $\sim 140^\circ\text{C}$  and the generated vapor was transported to the deposition zone through a dedicated delivery line using helium (He) as a carrier gas. For different  $\text{AlCl}_3$  flow, the soot deposition temperature was adjusted to maintain a good control over rare earth incorporation during the solution doping stage. Subsequently, the tube was removed from the MCVD lathe and soaked in a solution of methanol containing thulium chloride ( $\text{TmCl}_3 \cdot x\text{H}_2\text{O}$ , 99.9999%) and holmium chloride ( $\text{HoCl}_3 \cdot x\text{H}_2\text{O}$ , 99.999%). The dopant concentrations in the solution were adjusted for getting a specific Tm/Ho ratio in the fabricated preforms. Owing to the aqueous nature of the solution doping technique, a dehydration process was performed. Finally, the substrate tube was reassembled on the MCVD lathe for oxidation and sintering of the core layer. The tube was then collapsed into a solid rod in the usual manner. The preforms were post-processed to achieve a core diameter of  $\sim 8 \mu\text{m}$ . Additionally, the circular cladding was modified into a quasi-octagonal

shape for an enhanced pump absorption. The preforms were drawn into 200  $\mu\text{m}$  fibers and coated with a low index polymer.

The refractive index profile of the resulted fibers was measured with an optical fiber analyzer (IFA-100 from Interfiber Analysis, LLC). Electron disperse X-Ray spectroscopy (EDX) was used to identify the core composition. Figure 2 shows the dopants distribution of F-007 fiber. Absorption measurements were carried out using a white light source and an optical spectrum analyzer. The characteristics of the fabricated fibers are shown in Table 1. The  $\text{Tm}^{3+}$  concentration for fibers F-001 to F-004 is  $\sim 2$  wt%, while it is  $\sim 5$  wt% in fibers F-005 to F-008.



**Fig. 2.** Fiber F-007 dopants distribution.

**Table 1.** Tm and Tm:Ho co-doped fibers characteristics.

Fiber	NA	Core Abs at 790 nm [dB/m]	$\text{Tm}_2\text{O}_3$ [wt%]	$\text{Ho}_2\text{O}_3$ [wt%]	$\text{Al}_2\text{O}_3$ [wt%]
F-001	0.24	770	2.35	—	13.66
F-002	0.25	820	2.4	0.11	11.31
F-003	0.25	750	1.84	0.17	9.96
F-004	0.24	850	2.04	0.41	9.58
F-005	0.27	1670	6.44	—	11.65
F-006	0.29	1650	5.2	0.22	13.1
F-007	0.28	1530	6.32	0.56	10.24
F-008	0.29	1840	5.38	1.0	13.25

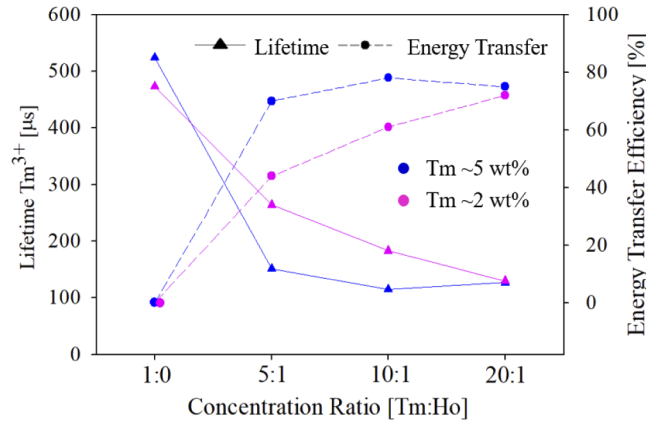
The fluorescence lifetime of the Tm and Tm:Ho co-doped fibers was analyzed to determine the  $\text{Tm}^{3+}$  to  $\text{Ho}^{3+}$  energy transfer, which can be observed by a shortening of the  $\text{Tm}^{3+}$  fluorescence decay time in the co-doped fibers. Lifetime from the  $^3\text{F}_4$  energy level manifold in thulium was measured under 790 nm pulsed pump light with a modulation frequency of 10 Hz and 10% duty cycle. An InGaAs photo-detector and a filter to suppress 2.1  $\mu\text{m}$  emission were used to capture fluorescence signal from thulium. The lifetime of the excited level was recorded on an oscilloscope. The efficiency of energy transfer from  $\text{Tm}^{3+}$  to  $\text{Ho}^{3+}$  ions was evaluated as [21]:

$$\eta = (\tau_{\text{Tm}} - \tau_{\text{Tm:Ho}}) / \tau_{\text{Tm}} \quad (1)$$

where,  $\tau_{\text{Tm}}$  is the lifetime of Tm ( $^3\text{F}_4$  manifold) in absence of holmium and  $\tau_{\text{Tm:Ho}}$  is the lifetime of Tm ( $^3\text{F}_4$  manifold) in the Tm:Ho co-doped system.

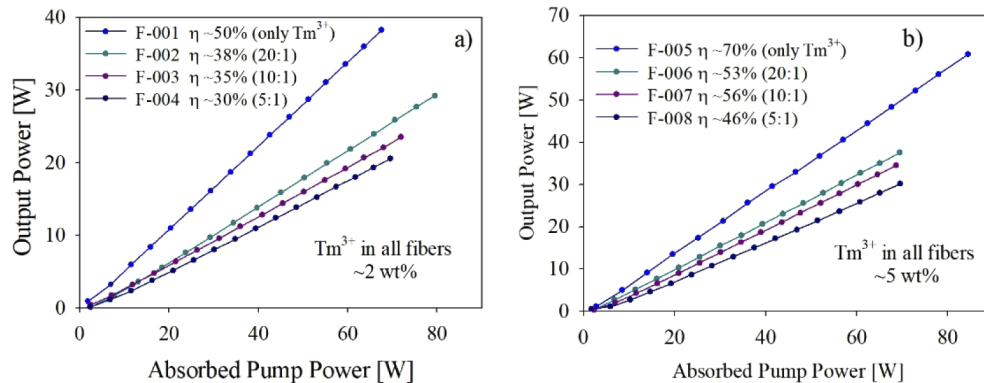
Figure 3 shows a significant reduction of up to 3.5 times in the fluorescence lifetime of  $\text{Tm}^{3+}$  in presence of  $\text{Ho}^{3+}$ . The change in fluorescence lifetime is a clear indication that the energy transfer process from donor ( $\text{Tm}^{3+}$ ) to acceptor ( $\text{Ho}^{3+}$ ) is occurring. Moreover, as shown in

Fig. 3, for Tm/Ho ratios of 5 and 10, a higher Tm<sup>3+</sup> concentration (Fibers: F-007 and F-008) has shown a much faster drop in Tm<sup>3+</sup> lifetime compared to the lower thulium concentration (Fiber: F-003 and F-004). This can be explained on the basis that an efficient two-for-one cross-relaxation process with a higher thulium concentration enables a greater number of excited ions available into the <sup>3</sup>F<sub>4</sub> level and hence the energy transfer process from Tm<sup>3+</sup> to Ho<sup>3+</sup> is more efficient at higher thulium concentration.



**Fig. 3.** Lifetime of thulium into the <sup>3</sup>F<sub>4</sub> energy level and the energy transfer efficiency as a function of Tm:Ho concentration ratio when pumped at 790 nm.

The laser performance of double clad fibers was measured in a 4% - 4% laser cavity. Pump light from a ~790 nm fiber coupled multimode laser diode was launched into the cladding through a combination of collimating lenses. A set of dichroic mirrors was used at both ends of the fiber to separate the pump and signal at the output. The slope efficiency was calculated adding the total output power from both ends of the fiber. The fiber length in all fibers was adjusted to obtain the maximum output power in a free-running laser configuration. The slope efficiencies of Tm:Ho co-doped fibers with ~2 wt% and ~5 wt% of Tm<sup>3+</sup> doping concentrations and different Tm/Ho ratios are presented in Figs. 4(a) and 4(b), respectively. The results of both graphs are summarized in Table 2.



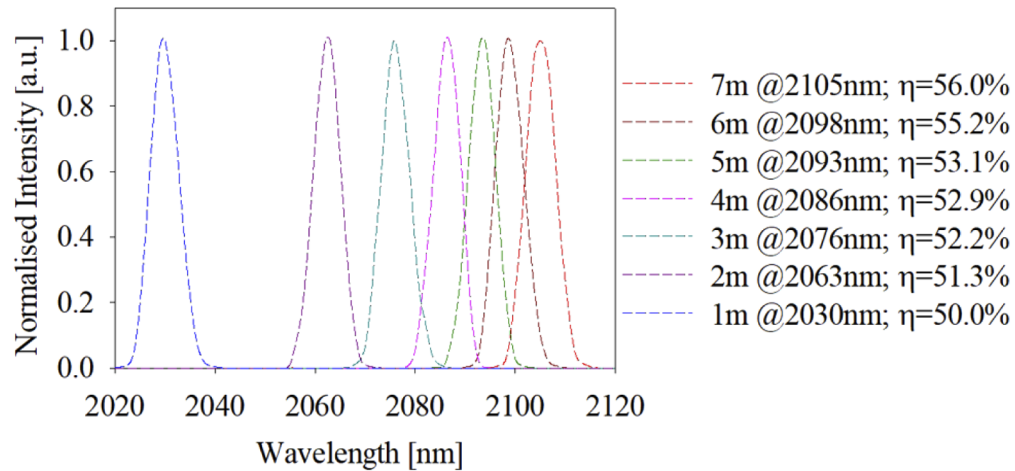
**Fig. 4.** Laser efficiency of Tm and Tm:Ho co-doped fibers w.r.t absorbed pump a) Tm<sup>3+</sup> ~2 wt% and b) Tm<sup>3+</sup> ~5 wt%.

**Table 2. Laser and energy transfer performance of Tm and Tm:Ho co-doped fibers**

Fiber	Tm:Ho Ratio	Length [m]	Emission [nm]	Laser Absorbed $\eta$ [%]	Tm $\rightarrow$ Ho ET $\eta$ [%]	Output Power [W]
F-001	—	12	2015	50	—	38.3
F-002	20:1	13	2090	38	72	29.6
F-003	10:1	15	2095	35	61	23.5
F-004	5:1	11	2103	30	44	20.6
F-005	—	8	2036	70	—	60.7
F-006	20:1	8	2082	53	75	34.5
F-007	10:1	7	2105	56	78	37.7
F-008	5:1	6	2114	46	70	30.1

Our results suggest that a better energy transfer efficiency occurs for Tm:Ho concentration ratios between 10:1 and 20:1. However, in order to achieve a maximum laser efficiency, the thulium concentration requires exceeding 2 wt% to promote an efficient two-for-one cross-relaxation process. The results of fibers containing a thulium concentration of  $\sim 5$  wt% support these observations. It is observed that when the Tm:Ho ratio is below 10:1, the laser efficiency suffers. This can be explained by the fact that as the holmium concentration in fiber increases, undesired energy transfer processes between Ho-Ho ion pairs will occur, which will then influence the laser efficiency.

Also, the performance of Tm:Ho fiber (F-007) lasers was analyzed as a function of fiber length and presented in Fig. 5. As the fiber length is shortened, the laser emission in a free running cavity is shifted towards the shorter wavelengths and has reached 2030 nm for 1 m long fiber, while still maintaining a good laser efficiency, which means the Tm:Ho co-doped fibers presented in this work has the potential for an extended wavelength of operation in the  $2.1 \mu\text{m}$  spectral region when diode pumped at  $79x \text{ nm}$  [15].

**Fig. 5.** Fiber F007 laser output spectrum as a function of fiber length.

### 3. Conclusion

In this paper, we have studied the  $\text{Tm}^{3+}$  ( $^3\text{F}_4$ ) to  $\text{Ho}^{3+}$  ( $^5\text{I}_7$ ) energy transfer and  $\text{Tm}^{3+}$  -  $\text{Tm}^{3+}$  cross-relaxation process as a function of the donor and acceptor concentrations in Tm–Ho co-doped aluminosilicate fibers. The fibers were diode pumped at 79x nm. Our study shows that the donor-acceptor concentration ratio that ranges from 10:1 to 20:1 and a thulium concentration of ~ 5 wt%, has resulted in the maximum laser efficiency for operation in the 2.1 microns wavelength region.

The hybrid gas phase-solution doping technique together with the MCVD process used to fabricate Tm–Ho co-doped fibers allows a more uniform distribution of the interacting rare earth ions across the core region. As a consequence, an efficient two-for-one cross-relaxation process in thulium followed by the energy transfer between thulium and holmium ions was obtained. Laser efficiency exceeding 55% at emission wavelengths of 2.1  $\mu\text{m}$  has been reached with possibility of achieving higher laser efficiency from further improvement in the donor acceptor energy transfer process.

All data supporting this study is available at <https://doi.org/10.5258/SOTON/D1264>

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### Disclosures

The authors declare no conflicts of interest.

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