光学学报

飞秒激光直写Tm∶YAP波导脉冲激光器

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摘要 利用飞秒激光直写技术制备Tm∶YAP光波导并实现1.9 μm波导调*Q*锁模激光输出。结合具有金属特性的$NbSe_2$薄膜作为可饱和吸收元件进行光学调制，波导激光输出的激光脉冲重复频率为7.8 GHz，最短脉宽为62 ps。这也是已报道的从Tm∶YAP波导中获得的最短激光脉宽。通过调整泵浦光的偏振，可以获得1855.87/1892.54 nm的双波长激光输出。结果表明，具有金属特性的$NbSe_2$薄膜在调制中红外超快脉冲激光器方面具有较大的应用价值。此外，双波长输出的紧凑型Tm∶YAP波导脉冲激光器在多功能集成光子学研究方面具有较好的应用前景。

关键词 激光器；飞秒激光直写；波导激光；Tm∶YAP；$NbSe_2$
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1 引 言

2 μm波段天然地具有“人眼安全”特性，在光学和光子学研究中扮演了重要的角色。同时，2 μm激光器也具有巨大的市场潜力，特别是在光探测和测距(LIDAR)、远程化学传感和直接光通信等自由空间光学研究中，2 μm激光器都具有广泛的应用，该领域的研究也受到人们越来越多的关注[1-4]。与钬离子(Ho^{3+})掺杂的固态激光器和激光二极管(例如基于GaSb的激光二极管)等光源相比，掺铥(Tm)激光器中三价铥离子(Tm^{3+})的主要吸收波段为780~800 nm，这可以通过利用技术较为成熟的商用激光二极管(例如基于AlGaAs的激光二极管)进行光学激发，使得掺Tm激光器在2 μm激光源的商业化方面处于领先地位[5-6]。固体激光器是常用的掺Tm激光系统，但二者均无法与集成光学技术兼容，这也在一定程度上限制了掺Tm激光器在实际应用中的发展。

波导激光技术是小型化固态激光器的有效解决方案之一，其核心增益介质为微米甚至纳米尺寸的光波导结构[7-11]。由于其紧凑的几何结构和对光场传输的有效限制，波导结构内光与物质相互作用的有效长度显著提升，有效地消除了光束发散，进而提高了光学增益，是研制小型激光器的理想平台[7-9]。此外，波导结构具有广泛的材料适用性和灵活的几何形貌，与多功能集成光子器件的兼容度较高[12]。在波导制备技术方面，与外延生长、载能离子束技术等相比，飞秒激光直写技术具有三维微纳加工能力，可以实现小于激光波长的单点扫描，在制备波导的灵活度和精细度方面具有一定优势，目前已经被运用到多种光学材料的波导制备中[10-11]。

基于Tm掺杂增益介质的连续波导激光已经在多种晶体和玻璃中实现，包括掺Tm石榴石(Tm∶YAG)晶体、掺Tm双钨酸钾(Tm∶KYW和Tm∶KLuW)晶体、掺Tm倍半氧化物(Tm∶Y_2O_3和Tm∶Lu_2O_3)晶体、掺Tm氟化物(Tm∶YLF)晶体和掺Tm氟化物玻璃(Tm∶ZBLAN)等[7-9]。然而，有关Tm脉冲波导激光的报道较少，这与所制备的波导质量有一定的关系。掺有Tm^{3+}离子的铝酸钇(Tm∶YAP)晶体是波导激光器的理想增益介质。与Tm∶YAG相比，Tm∶YAP具有较大的吸收和发射截面以及吸收带宽，更适合研制低阈值激光器件[8]。此外，YAP本身的光学各向异性允许其在具有不同偏振的光泵浦下对输出激光的性能特征进行微调，这也在一定程度上提升了激光器的灵活度，增强了其功能性。然而，目前有关Tm∶YAP波导激光的报道相当匮乏。

波导脉冲激光器可以通过主动/被动调*Q*或锁模

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技术来实现。与需要外部信号进行光学调制的主动调制方法相比，被动调制手段通常依赖于放置在激光腔内的可饱和吸收（SA）元件，这与需要紧凑型封装的小型化激光器设计理念更加匹配[9]。SA 元件在初始化和稳定脉冲激光产生方面起着至关重要的作用，这也一直驱动着人们寻找理想的 SA 材料。二维过渡金属二硫化物（TMDC）是一类新兴材料，因具有高损伤阈值、超快响应、宽带吸收等特性，成为极具吸引力的 SA 元件候选材料[13-15]。其中，以 MoS_2、$MoSe_2$、WS_2、WSe_2 等为代表的半导体 TMDC 具有与层堆叠数相关的光电性质，已被广泛应用于光电子学研究中[16-18]。相比之下，关于 NbS_2 和 $NbSe_2$ 等具有金属特性的 TMDC 的研究相对匮乏。该类 TMDC 在近红外到远红外波段的超宽范围内具有良好的光学响应，且其光电性质与层堆叠数无关，在宽带光调制应用方面具有较大的潜力[19]。

本文利用飞秒激光直写技术制备了 Tm∶YAP 包层光波导，结合基于 $NbSe_2$ 二维材料薄膜的 SA 元件，实现了重复频率为 7.8 GHz、锁模脉宽为 62 ps 的 1856/1893 nm 双波长波导调 Q 锁模激光输出。实验结果表明：1）飞秒激光直写 Tm∶YAP 包层光波导在小型化 2 μm 激光光源中具有一定的应用潜力；2）$NbSe_2$ 具备在 2 μm 波段调制产生高重复频率波导脉冲激光的能力。该工作为研制新型可集成 2 μm 脉冲激光光源提供了一定的思路。

2 波导和 SA 体材料的制备与表征

2.1 Tm∶YAP 包层波导和 $NbSe_2$ 薄膜的制备

利用飞秒激光直写技术在 Tm∶YAP 晶体［Tm^{3+} 掺杂浓度（原子数分数）为 3%，晶体切向为 c 切，晶体尺寸为 12 mm（a）×10 mm（b）×2 mm（c），晶体表面已作光学抛光处理］中制备直径为 50 μm 的圆形包层波导，其端面显微镜图像如图 1（a）所示。制备过程中使用的飞秒激光器（Astrella，Coherent Inc.，USA）中心波长为 800 nm，激光通过一个显微物镜（40×，数值孔径 NA=0.65）聚焦在晶体表面以下 190 μm 处。经过对飞秒激光加工参数的优化（通过测试波导损耗对加工参数进行优化），设置飞秒激光脉冲的重复频率为 1 kHz，脉宽为 38 fs，脉冲能量为 0.13 μJ。在制备加工过程中，将 Tm∶YAP 晶体放置在由 PC 控制的三维平移台上，平移台的移动速度固定为 0.5 mm/s。本实验中使用的 $NbSe_2$ 二维材料薄膜样品由化学气相沉积（CVD）方法合成，薄膜样品由六碳科技有限公司（6Carbon Technology，China）提供。

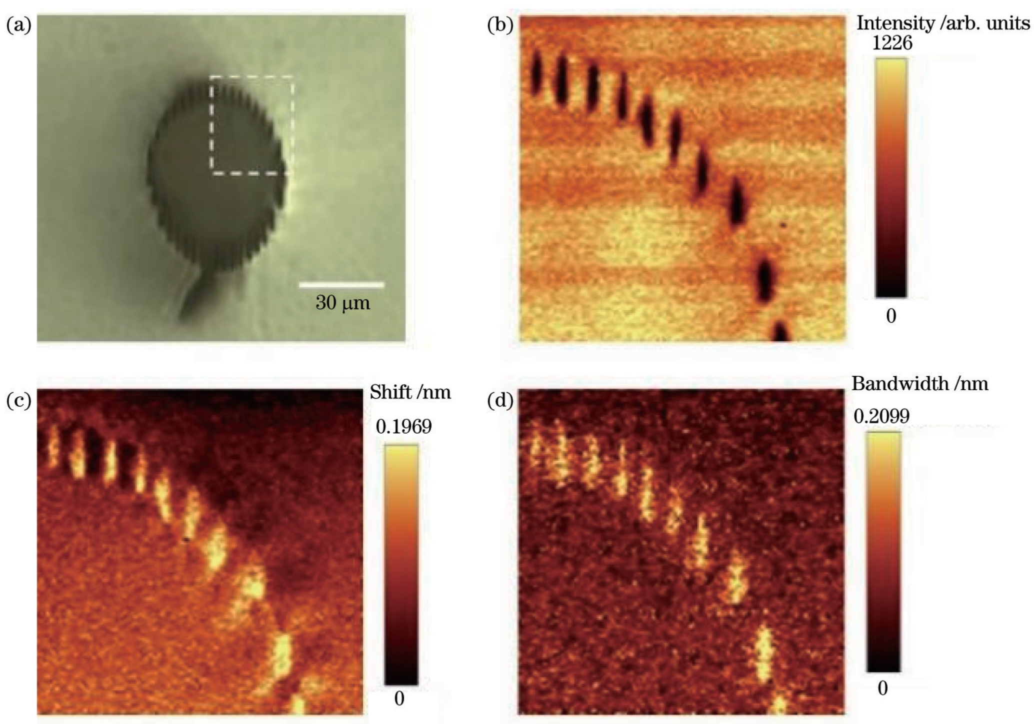

图 1 飞秒激光直写 Tm∶YAP 波导。（a）飞秒激光直写 Tm∶YAP 波导端面显微镜图像；共聚焦微荧光发射线的（b）强度、（c）频移和（d）带宽的二维空间分布图，测量区域为显微镜图像中虚线框区域

Fig. 1 Femtosecond laser direct writing of Tm∶YAP waveguide. (a) Optical transmission micrograph of cladding waveguide fabricated in Tm∶YAP crystal; spatial 2D distributions of micro-photoluminescence (μ-PL) (b) intensity, (c) shift, and (d) bandwidth obtained from the cladding waveguide cross section, and the μ-PL analysis is conducted in the dashed frame in Fig. 1(a)

2.2 飞秒激光直写波导局域晶格损伤分析

在室温下使用共聚焦微荧光显微镜（Alpha300 R，WITec GmBH）对 Tm∶YAP 中飞秒激光直写的损伤微区（即波导包层区域）进行测试，进而研究飞秒激

光脉冲对局域晶格损伤情况和波导内荧光性质的保留情况，实验结果如图 1(b)～(d)所示。在实验中，利用一束波长为 488 nm 的连续激光作为荧光激发光源，通过一个 100× 的显微物镜（*NA*=0.9）聚焦在样品端面，其发射的信号通过一个光谱仪（UHTS 300 SMFC VIS）进行收集，并利用 WITec 软件对微荧光测试结果进行处理分析。可以看到，相较于未被加工的体材料区域，飞秒激光聚焦微区的光谱强度有一定降低，频率红移，并且伴随光谱红移而展宽。这些现象说明飞秒激光加工在局部区域产生了晶格扭曲和损伤，这也导致了加工微区的折射率发生变化[11]。波导核心区域的荧光性质几乎没有变化，说明原有晶体的荧光性质在波导区域得到很好的保留，有利于波导激光的产生。

2.3 波导的导波性能和 $NbSe_2$ 薄膜饱和吸收性表征

为了对波导的导波性能进行研究，采用偏振可调的 1550 nm 连续激光作为波导激励光源，通过一个焦距为 25 mm 的平凸透镜耦合进波导，再通过一个 20× 显微物镜（*NA*=0.4）对出射光进行收集。图 2(a)所示为 Tm∶YAP 包层波导在不同角度偏振下的透过率，可以看到，波导在 TM 偏振下的透过率略高于 TE 偏振下的透过率。实验测得在 TM 和 TE 偏振下波导的插入损耗分别约为 1.21 dB 和 1.98 dB，这是因为波导截面几何形状在横向的尺寸相比于竖向更小，较为粗糙的波导边界对导波区域光传输的影响更大，所产生的散射损耗也更大。利用连续钛宝石激光器（Coherent MBR-PE）代替 1550 nm 激光器作为泵浦光源，通过调节泵浦光波长（793.7～816.1 nm）对波导的激光出射情况进行测试。在激光测试中，波导的两个端面分别紧紧贴合泵浦腔镜 M1（两面分别镀有在 780～980 nm 透过率＞99.8% 的增透膜及在 1850～1950 nm 反射率＞99.9% 的高反射膜）和输出腔镜 M2（两面分别镀有在 1700～2700 nm 反射率＞99.5% 的高反膜及在 1700～2700 nm 反射率约为 40% 的部分反射膜），构成激光谐振腔。实验中，利用 900 nm 长通滤波片滤掉输出激光中残余的泵浦光。在保证泵浦光功率一定的情况下改变泵浦光波长，得到随泵浦光波长变化的输出激光相对强度，如图 2(b)所示。结果显示，所制备的 Tm∶YAP 包层波导在 793.7～816.1 nm 光的泵浦下都能实现激光输出，在 795.7 nm、799.3 nm 和 802 nm 处出射光功率较大。这样的宽吸收特性有利于波导与二极管激光器泵浦兼容，可以很好地克服二极管激光器功率提升时出现的波长偏移问题。

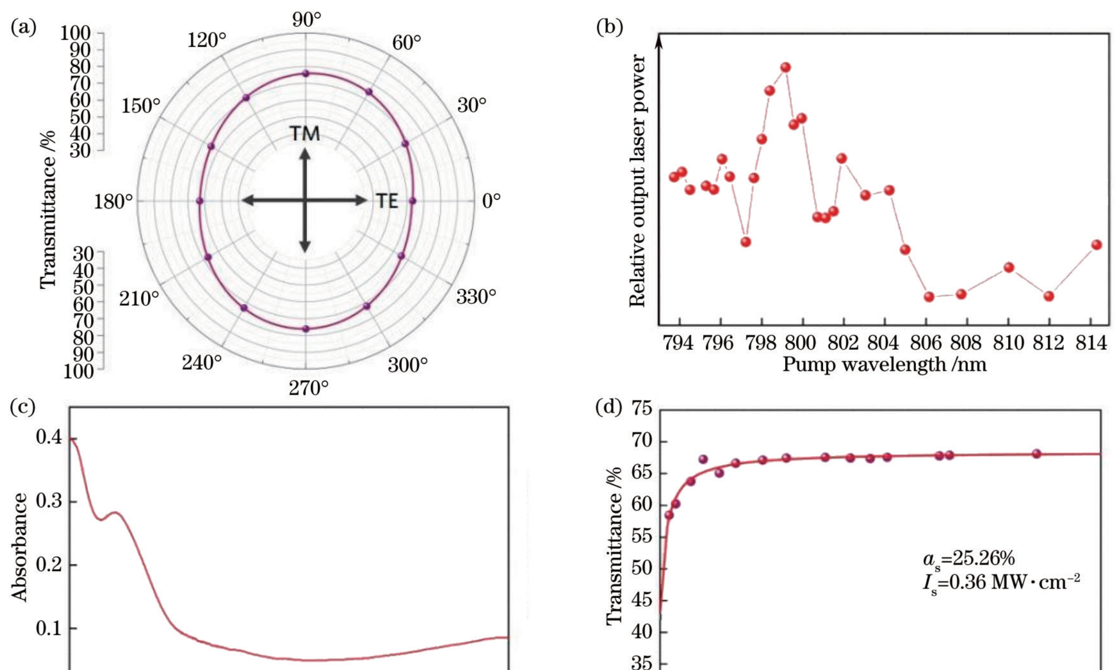

图 2 Tm∶YAP 包层波导和 $NbSe_2$ 薄膜样品的光学性能。(a) Tm∶YAP 包层波导在不同偏振下的透过率（波长为 1550 nm）；(b)不同波长泵浦光激励下出射激光的相对强度；(c) $NbSe_2$ 薄膜样品的线性吸收谱及(d)其在 2 μm 下 *I* 扫描测试结果

Fig. 2 Optical properties of Tm∶YAP cladding waveguide and $NbSe_2$ thin film. (a) Transmittance of fabricated Tm∶YAP cladding waveguide in different polarizations; (b) relative output laser power under optical pumping at different wavelengths; (c) linear absorption spectrum and (d) 2 μm *I*-scanning measurement result of $NbSe_2$ thin film

利用紫外/可见/红外分光光度计（Cary 5000 UV-Vis-NIR，Agilent Technologies）对 $NbSe_2$ 薄膜样品的线性光学吸收性能进行测试，结果如图 2(c)所示。可以看到，$NbSe_2$ 薄膜样品具有较宽的吸收光谱，说明

$NbSe_2$具有较宽的光学响应范围，有望实现超宽波段的SA应用。为了对$NbSe_2$薄膜的SA性能进行测试，进行I扫描实验。利用波长为1970 nm的飞秒光纤激光器（Rainbow 2000，NPI Lasers）作为测试光源，其激光脉宽为200 fs，重复频率为80 MHz。光源出射激光经过焦距为25 mm和75 mm的两个平凸透镜进行聚焦，将样品放置在焦点处，焦点处光斑半径约为80 μm，光源功率通过一个连续衰减片进行调整。如图2（d）所示，$NbSe_2$薄膜在1970 nm的透过率随着光强的提升呈现饱和趋势，通过数据拟合得到其调制深度为25.26%，饱和强度为0.36 MW/cm^2。该结果显示$NbSe_2$薄膜可应用到2 μm脉冲激光调制中。

3　Tm∶YAP脉冲波导激光

将$NbSe_2$薄膜作为SA元件插到波导端面和出射激光腔镜中间，并且使其紧贴着波导出射端面和出射腔镜，保证与波导模式充分作用。根据先前测试的连续激光结果[图2（b）]，将泵浦光波长固定在799.3 nm来激励Tm∶YAP波导，通过调节泵浦光功率，成功得到重复频率为7.8 GHz的双波长Tm∶YAP调Q锁模波导激光输出（图3）。其中，输出激光脉冲由2 μm InGaAs光电探测器（ET-5000）和25 GHz宽带实时数字示波器（Tektronix，MSO 72504DX）记录，出射激光的功率和模式分别由光电二极管锗探头（Thorlabs S122C）和光束质量分析仪（DataRay's Beam'R2）收集测量，输出激光的光谱通过光纤光谱仪（NIRQUEST256-2.5，900～2500 nm）收集测量。如图3（a）所示，所制备的Tm∶YAP在TE（TM）偏振光的泵浦下激光输出阈值约为45 mW（43 mW），最大输出功率为65 mW（34 mW），斜效率为11.86%（6.02%）。在TE偏振光泵浦下可获得更优的激光性能，主要是因为体材料在该晶体方向具有更高的激光增益。实验中，通过调整泵浦光的偏振，可以实现对输出波导脉冲激光波长的调整，获得1855.87 nm和1892.54 nm的双波长激光输出，该特性能够应用于输出波长可调的小型激光光源。与Tm∶YAP连续波导输出激光相比，经过$NbSe_2$薄膜SA元件调制的波导脉冲激光的阈值较高（连续激光模式下激光阈值约为30 mW）、斜效率和最大输出功率更低（连续激光模式下激光最大输出功率＞150 mW，斜效率约为25.54%），这主要是因为$NbSe_2$薄膜的插入引入了一定的光学吸收损耗，在一定程度上降低了波导激光腔的光学增益。在输出波导激光模式、激光波长以及偏振依赖特性方面，脉冲模式下的波导输出激光与连续

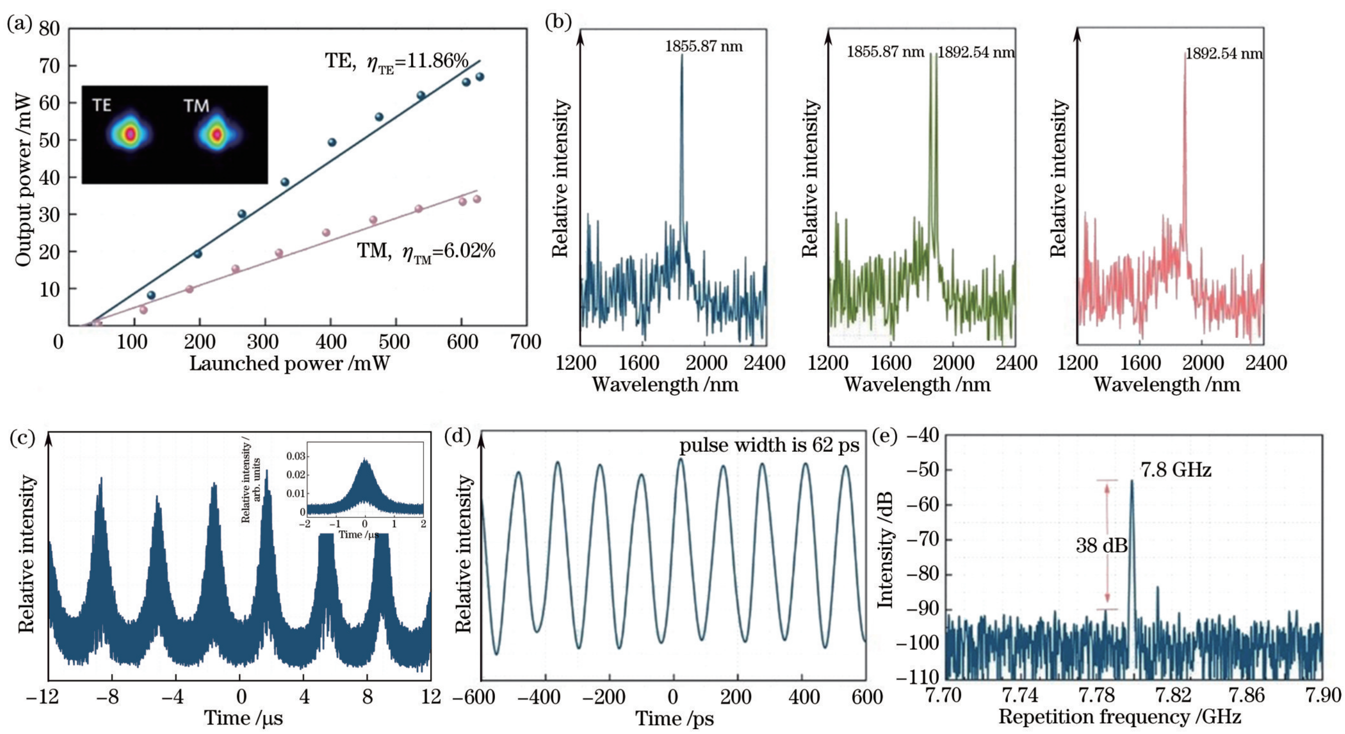

图3　Tm∶YAP调Q锁模波导激光性能。（a）在TE和TM偏振下的出射激光功率随泵浦功率的变化关系，插图为两种偏振下的近场模式分布图；（b）出射激光波长随泵浦光偏振变化的调谐图（偏振方向从左到右分别为TE、45°、TM）；（c）调Q包络序列图，插图所示为单个调Q包络；（d）锁模序列；（e）出射激光频谱图

Fig. 3　Laser performance of Tm∶YAP Q-switched mode-locked waveguide. (a) Relationship between output laser power and pump power under TE and TM polarizations, the insets show the near-field modal profiles under two different polarizations; (b) wavelength tuning of laser emission under optical pumping with different polarizations (TE, 45°, and TM from left to right); (c) Q-switched envelope sequences, and the inset shows a single Q-switched envelope; (d) mode-locked trains; (e) RF spectrum

模式下的输出激光特性相近，说明 $NbSe_2$ 薄膜的插入对光波导导波特性的影响较小。波导脉冲激光的调 Q 包络序列和锁模序列分别如图 3(c)、(d)所示，可以确定锁模脉宽约为 62 ps，出射激光的重复频率为 7.8 GHz，这是目前已报道的脉宽最窄的 Tm：YAP 波导脉冲激光。在 600 mW 泵浦功率的激励下，测量记录了出射激光性能稳定情况，发现激光功率在 1 h 内的变化小于 10%，所获得的包络脉冲不一致性小于 5%，说明所制备的波导结构以及设计的波导谐振腔具有较好的热稳定性。进一步的激光稳定性提升需要对波导制备参数、激光光腔的散热等进行优化，进而获得高品质、低损耗的波导增益介质以及高质量、高稳定的激光光腔。在高功率下，由于 SA 元件的热效应，波形出现轻微扰动，这对于输出激光的稳定性也有影响。实验结束时 SA 元件并未发生损坏，表明 $NbSe_2$ 薄膜材料具有较高的抗损伤阈值。

在本波导激光实验中，通过对泵浦条件和波导耦合条件的调节，未能实现稳定状态的连续锁模激光输出，这主要是因为本实验中的波导激光腔缺乏有效的色散调控。事实上，在紧凑的波导激光腔中很难再插入色散调控元件。为了保证波导激光腔紧凑型的同时实现有效的色散调控，可以通过调整波导端面和腔镜或 SA 元件之间的薄空气间隙实现干涉色散控制。在该操作中，充满空气的间隙可等效为 Gires-Tournois 标准具结构，能够提供足够的色散管理来实现连续锁模激光输出[9,20]。这是本课题组下一步的研究内容。

4 结　　论

利用飞秒激光直写技术在 Tm：YAP 晶体中制备了直径约为 50 μm 的包层波导，在 793.7～816.1 nm 光的泵浦下都支持激光产生。利用 CVD 方法制备了 $NbSe_2$ 薄膜材料，测得其在 2 μm 处的调制深度和饱和强度分别为 25.26% 和 0.36 MW/cm^2。将其作为 SA 元件，得到重复频率为 7.8 GHz，锁模脉宽约为 62 ps 的 1855.87/1892.54 nm 双波长可调 Tm：YAP 调 Q 锁模波导激光输出，这是目前已报道的脉宽最窄的 Tm：YAP 波导脉冲激光，为多功能、紧凑型波导光源的制备提供了实验依据。此外，还展示了 $NbSe_2$ 薄膜材料在 2 μm 波段调制产生高重复频率、窄脉宽波导激光的能力，其有望作为 SA 体材料在更远波段发挥调制作用。

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Tm∶YAP Waveguide Pulsed Lasers of Femtosecond Laser Direct Writing

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Abstract

Objective The eye-safe nature of 2 μm lasers endow them with great market potential, especially in free space applications, such as light detection and ranging (LIDAR), remote chemical sensing, and direct optical communication. Accordingly, there is an increasing research interest in 2 μm lasers operating in both continuous-wave (CW) and pulsed regimes. Fiber lasers and bulk lasers are the most commonly employed Tm-laser systems, while neither of them is compatible with integrated photonics, which hinders the development of Tm lasers in practical applications. A compact and robust solution to the minimization of solid-state lasers is a waveguide laser, in which a well-structured optical waveguide is utilized as the gain medium and the key component of the laser cavity. Due to their compact geometries, waveguide structures offer strong confinement of light propagation over a relatively long interaction length, effectively removing the beam divergence and in turn enhancing the optical gain.

As one of the most attractive gain media for 2 μm solid-state lasers, Tm∶YAP has relatively higher absorption and emission cross sections and a relatively broad absorption band. However, Tm∶YAP waveguide lasers operating either in CW or pulsed regimes have not been demonstrated till now. Waveguide lasers operating in pulsed regimes, typically by *Q*-switching or mode locking techniques, can be realized by both active and passive methods. In contrast to active methods that require external signals for optical modulation, passive methods typically rely on the saturable absorber (SA) elements placed inside the laser cavity for light self-modulation, allowing for robust laser designs with compact packages. We aim to fabricate high-quality Tm∶YAP waveguides and experimentally demonstrate 2 μm waveguide lasers operating at pulsed regimes.

Methods We adopt the femtosecond laser direct writing (FsLDW) technique for fabricating cladding waveguides in a Tm∶YAP crystal wafer with doping concentration of 3% (atomic number fraction) Tm^{3+} ions and dimensions of 12 mm (*a*)× 10 mm (*b*) × 2 mm (*c*), and the surface of the crystal wafer is polished to the optical-grade quality. The crystal wafer is placed on a PC-controlled *XYZ* micro-position stage for precise translation with a constant velocity of 0.5 mm/s. The 800 nm laser is delivered by a femtosecond laser system with a pulse width of 38 fs and repetition rate of 1 kHz, and focused by a microscope objective lens beneath the largest crystal surface. A pulsed energy of 0.13 μJ is set for producing laser-damage tracks and avoiding crystal cracking. For each scanning, a damage track induced by an fs-laser with a vertical length of 10 μm is produced, and a cladding waveguide with a diameter of around 50 μm in the crystal is obtained after multiple scannings. The main intention of choosing this FsLDW parameter combination is to realize low-loss waveguides with optimized guiding and lasing performances. The employed $NbSe_2$ thin film SA element is prepared by chemical vapor deposition (CVD) method.

Results and Discussions To study the FsLDW-induced crystalline lattice changes and the preservation of fluorescence properties within waveguide volume, we conduct micro-photoluminescence (μ-PL) analysis by employing a fiber confocal microscope at room temperature. In the experiment, a CW 488 nm laser source for luminescence excitation is focused through the cladding waveguide cross-section with a depth of 10 μm by a microscope objective, and the emitted signal is detected via a spectrometer. The μ-PL intensity collected from the waveguide and the bulk material has slight differences, which results in very slight fluorescence quenching in the guiding area due to the lattice damage caused by fs-laser pulses. The nearly identical intensity indicates sound preservation of the original luminescence properties of Tm∶YAP crystal in the guiding area. There is a noticeable intensity reduction of μ-PL emission in the filament region compared with the bulk region, which indicates the partial lattice distortion and damage in the laser-modified region.

By inserting the $NbSe_2$ thin film as an SA element between the end face of the waveguide and the laser cavity mirror, we successfully obtain a waveguide pulsed laser with a repetition rate of 7.8 GHz under optical pumping at 799.3 nm. The lasing threshold of the prepared Tm∶YAP waveguide is about 45 mW (43 mW) under the optical pumping with TE (TM) polarization. Correspondingly, the maximum output power is 65 mW (34 mW) and the slope efficiency is 11.86% (6.02%). The superior lasing performance under TE polarization is mainly due to the higher lasing gain of the bulk material along this crystal direction. In the experiments, by adjusting the polarization of the pump light, the wavelength of the output waveguide pulse laser can be adjusted, and dual-wavelength laser output of 1855.87 nm and 1892.54 nm can be obtained. Compared with Tm∶YAP continuous waveguide laser, the waveguide pulsed laser modulated by $NbSe_2$ thin film SA element has a higher threshold, lower laser slope efficiency, and maximum output power. This is mainly because the insertion of the $NbSe_2$ thin film introduces a certain optical absorption loss, reducing the optical gain of the waveguide laser cavity to a certain extent. In terms of output waveguide laser mode, laser wavelength, and polarization dependence characteristics, the waveguide laser in pulsed mode is similar to that in the continuous wave mode. This shows that the insertion of $NbSe_2$ thin films exerts little influence on the waveguide wave characteristics of the optical waveguide. The experimentally determined mode-locked pulse width is about 62 ps and the repetition frequency of the outgoing laser is 7.8 GHz. The obtained Tm∶YAP waveguide pulsed laser has the narrowest pulse till now.

Conclusions The demonstration of a 1.9 μm *Q*-switched mode-locked Tm∶YAP cladding waveguide laser fabricated by FsLDW is reported. Modulated by metallic $NbSe_2$ thin films as an SA element, the fabricated waveguide laser delivers laser pulses has a pulse duration of as short as 62 ps at a fundamental repetition rate of up to 7.8 GHz. This is up-to-date with the shortest laser pulses that are achieved from Tm∶YAP waveguides. By adjusting the polarizations of the optical pumping, a dual-wavelength laser operating at 1855.87/1892.54 nm is obtained. The results indicate promising applications of metallic $NbSe_2$ thin films for modulating mid-infrared ultra-fast pulsed lasers and compact Tm∶YAP waveguide lasers for multi-functional integrated photonics.

Key words lasers; femtosecond laser direct writing; waveguide laser; Tm∶YAP; $NbSe_2$