

Supporting Information

The Five-Photon Absorption Upconversion Lasing from on-Chip Whisper-gallery Mode

Ying Huang^{1,#}, Hai Zhu^{1,*,#}, Huiying Zheng¹, Ziyang Tang¹, Jianwen Dong,¹ Shichen Su,²
Yan Shen,³ Xuchun Gui,³ Shaozhi Deng,³ and Zikang Tang⁴

¹State Key Laboratory of Optoelectronic Materials and Technologies, School of Physics, Sun Yat-Sen University, Guangzhou 510275, China

²Institute of Optoelectronic Material and Technology, South China Normal University, Guangzhou 510631, China.

³State Key Laboratory of Optoelectronic Materials and Technologies, School of Electronics and Information Technology, Sun Yat-Sen University, Guangzhou 510275, China

⁴The Institute of Applied Physics and Materials Engineering, University of Macau, Avenida da Universidade, Taipa, Macau, China

#The authors contributed equally to this work.

E-mail: zhuhai5@mail.sysu.edu.cn

In order to demonstrate the excellent nonlinear optical properties of single-crystal film at room-temperature (RT), the low-dimensional quantum structure was designed. Based on the artificial fabricated quantum microstructure, the upconversion lasing can be tunable. In experiment, we have realized the RT upconversion lasing based on $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$ multiple quantum well (MQW) through 1900 nm excitation (**Fig. S1**). It is noted that the lasing peak of ZnO MQW emerges a blue shift obviously compare with bare ZnO film. The detail upconversion lasing spectra versus the different pumping power (with excitation light 1900 nm) was present (**Fig. S2**).

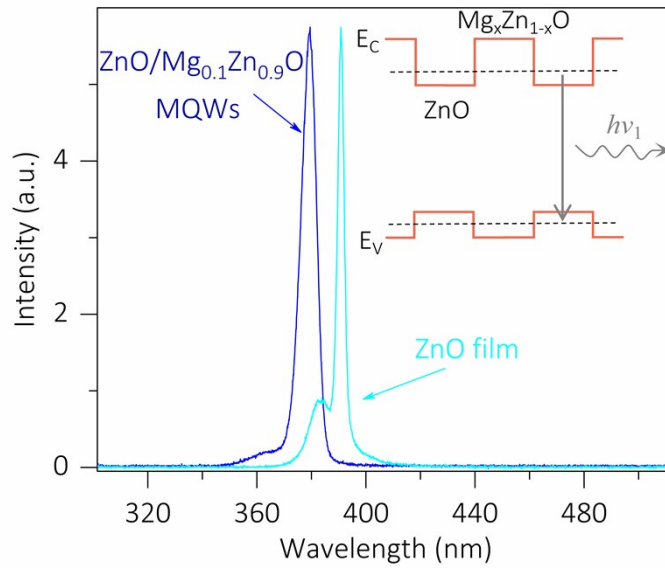


Figure S1. The optical properties comparison between film and quantum structure. The upconversion lasing spectra from bare ZnO film and $\text{Mg}_{0.1}\text{Zn}_{0.9}\text{O}/\text{ZnO}$ multiple quantum well at RT respectively. Inset, the schematic diagram of $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$ QW energy band and the transition between the sublevels.

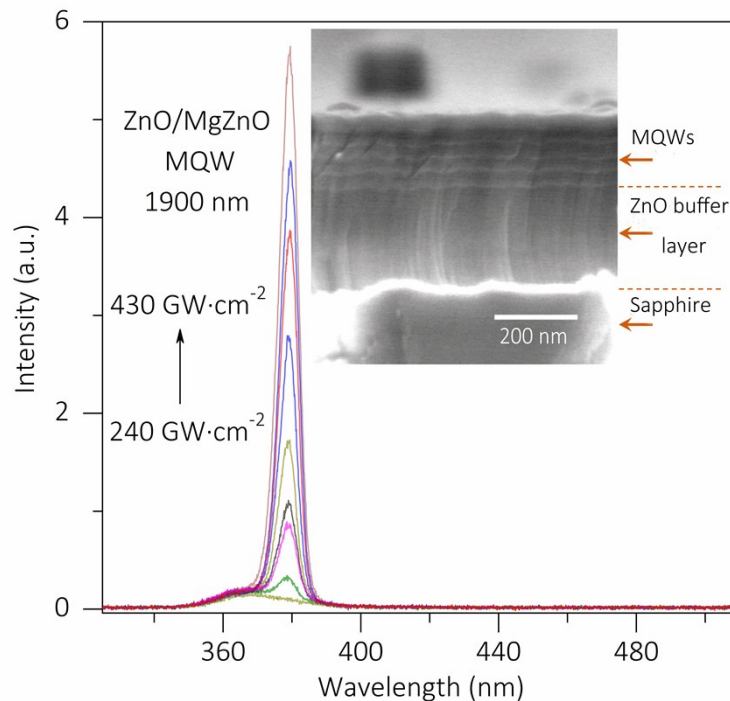


Figure S2. Upconversion lasing in ZnO-based QW. Detailed upconversion lasing spectra from MgZnO/ZnO multiple quantum well at RT. Inset, the cross-section SEM image of $\text{Mg}_x\text{Zn}_{1-x}\text{O}/\text{ZnO}$ QW structure that epitaxy on sapphire substrate.

Figure S3a depicts the detail evolution of four-photon (1500 nm) excited upconversion emission spectra with a progressively increasing of pump density. Furthermore, the intensity of NBE and FWHM of upconversion lasing (THG) as a function of pumping density are shown in **Figure S3b**, and c, respectively. However, the intensity of THG peak versus pumping power exhibits a cubic function and its FWHM shows no obvious changing. Above results confirm the THG peak is originated from the third-order nonlinear optical process through the virtual states.

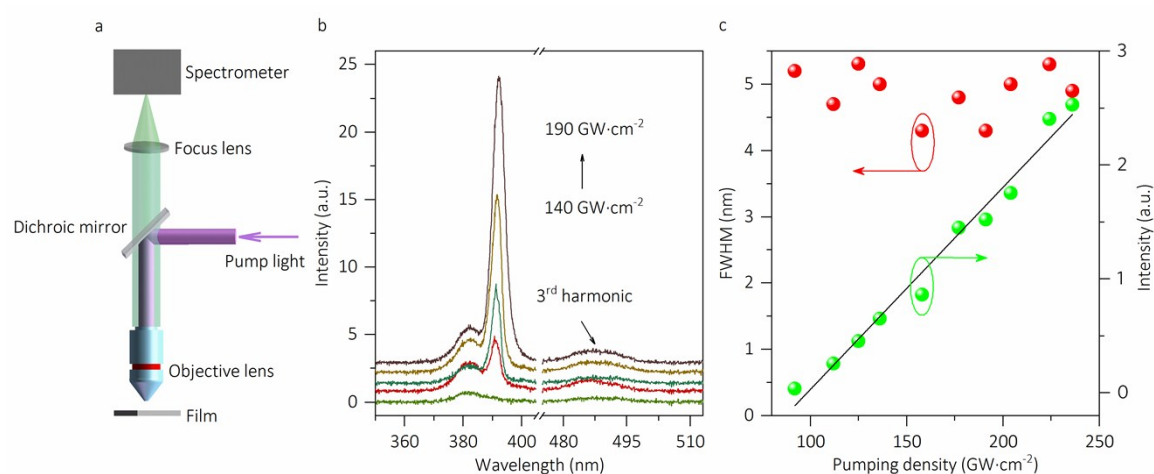


Figure S3. Experimental set-up of PL and the third-harmonic peaks of the pumping laser. **a** the experimental set-up of femtosecond laser pumping system for MPA upconversion fluorescence. **b** The NBE located at 390 nm was realized through 4PA simultaneous. The Pumping density was tuned from 140 GW·cm⁻² to 190 GW·cm⁻² with a temporal width (repetition rate) of 50 fs (1 kHz) and the third harmonic peaks of the tunable pumping light were also indicated obviously in the spectra. **c** The emission intensity and FWHM of THG peak versus pumping intensity at RT.

In addition, the hexagonal structure resonator was fabricated as shown by the SEM image of **Fig. S4a**. Here, the microcavity with a diameter of 20 μm was excited (1900 nm) under different excitation power. The **figure. S4 b-f** display the near-field emission pattern of the 5PA upconversion WGM lasing in the microcavity under different pumping power. Remarkable, the leakage lasing radiation light can be observed from six direction of hexagonal cavity clearly.

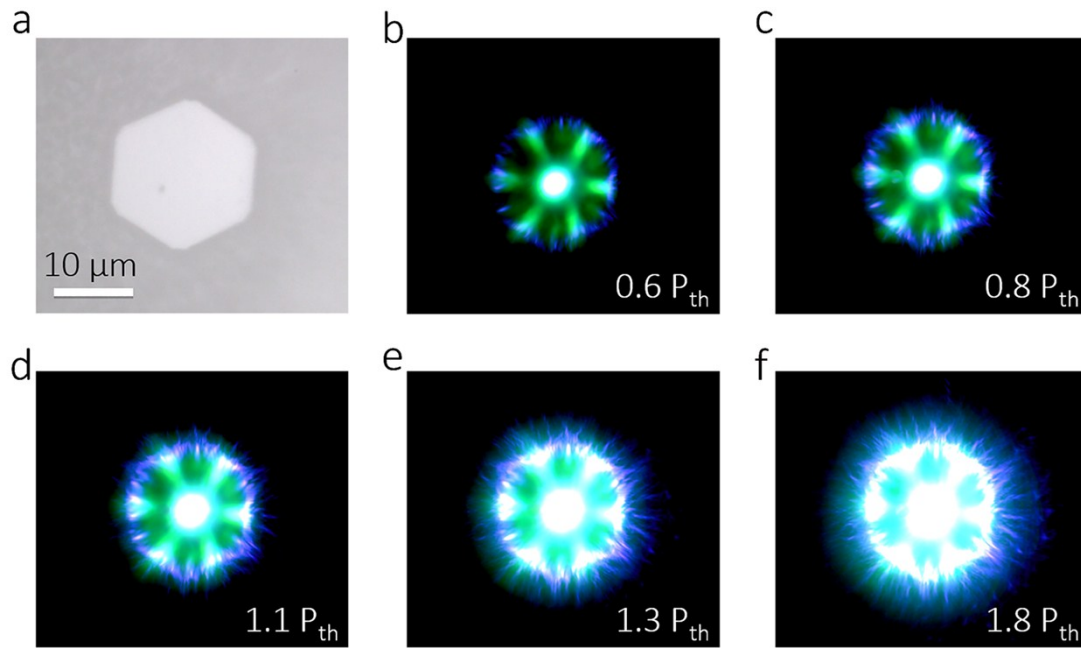


Figure S4. The near-field emission pattern of WGM lasing through 5PA in hexagonal structure cavity. **a** The SEM image shows the morphology of the fabricated hexagonal microcavity. Here, the cavity with a diameter of 20 μm was excited (1900 nm) under different excitation power. **b-d** The near-field emission pattern of device that under series pumping power as 0.6 P_{th}, 0.8 P_{th}, 1.1 P_{th}, 1.3 P_{th}, 1.8 P_{th} respectively.