Supporting Information

Transfroming of Random Lasing to Fabry–Perot Lasing: Observation of High Temperature Lasing from Carbon Dots

Yiqun Ni[a,b], Xiangyang Li[a], Wenqing Liang[a], Shaofeng Zhang[a], Xuesong Xu[a], Zhenyuan Li[a], Ling Li[a], Yonghong Shao[b], Shuangchen Ruan[a], and Wenfei Zhang[a]*

[a] Shenzhen Key Laboratory of Laser Engineering, College of Physics and Optoelectronic Engineering, Shenzhen University, Shenzhen 518060, P. R. China

[b] Key Laboratory of Optoelectronic Devices and Systems of Ministry of Education and Guangdong Province, College of Physics and Optoelectronic Engineering, Shenzhen University, Shenzhen, 518060, China

* Corresponding author: zhangwf@szu.edu.cn

Key words: Carbon dots, Red emission, High temperature Fabry–Perot lasing

Experimental Section:

Carbon dots fabrication:

In a typical procedure, 1,3-Dihydroxynaphthalene (10 mg, carbon source) and sulphuric acid (3 ml, oxidant) were dissolved in ethanol (10 mL) to form a clear solution under ultrasonication for 10 min. Then the solution was transferred into poly(tetrafluoroethylene) (Teflon)-lined autoclave (50ml) and placed in oven at 190 ºC for 3 hours. After the reaction, the reactor was cooled down to room temperature naturally. The obtained reaction mixture was purified via silica column chromatography using a mixture of methyl alcohol and dichloromethane as the eluent. In this process, the volume ratio of dichloromethane and methyl alcohol is set to 50:7. Finally, Y/R/D-CDs solutions with yellow, red, and deep red emission were obtained after the above fabrication processes.

Laser cavity fabrication:

Firstly, the as prepared R-CDs were mixed with epoxy under continuous stirring to form 0.5% (weight ratio) R-CDs/epoxy composite. Secondly, the R-CDs/epoxy composite was spin coated on an Al mirror at 2000 rpm for 10 seconds to form R-CDs/epoxy composite gain layer. Simultaneously, the R-CDs/epoxy composite layer was cured into a solid film under 60 ºC for 1 hour. At last, a quartz plate was covered on the top of the R-CDs/epoxy composite gain film to form a planar Fabry-Perot microcavity.

Characterization method:

Morphology and height of three types of CDs were investigated by a JOEL JEM-2100F high-resolution transmission electron microscope (HR-TEM) and Bruker Dimension Icon (AFM) respectively. The structure of CDs was analyzed by a Horiba-JY T64000 Raman system and MalvernPanalytical Empyrean (XDR). Surface functional groups of CDs were studied by Thermo Scientific K-Alpha X-ray Photoelectron Spectrometer (XPS) and *Bruker Vertex* 80V Fourier transform infrared (FTIR) spectroscopy. Fluorescence spectra, excitation spectra, UV−vis absorption spectra, resolved fluorescence lifetimes and quantum efficiency were measured by Spectrofluorometer FS5 (Edinburgh instruments).

The lasing characteristics of the Fabry-Perot microcavity laser were studied under optical excitation by a neodymium-doped yttrium aluminum garnet (Nd:YAG) pulsed laser (355 nm, 10 Hz, Continuum Surelite, San Jose, CA, USA)and an optical parameter oscillator (Continuum Horizon, San Jose, CA, USA). A 532nm pump beam with a diameter of 100 μm was focused on the gain film through a lens. Then the light emission from the cavity was coupled into a HORIBA iHR320 spectrometer (Minami-ku, Kyoto, Japan) via an optical fiber. Gain of the film was measured by utilizing variable stripe length method. For elevated temperature measurement, the optical gain and lasing measurement were both operated in a temperature controllable stage (linkam HFS-600).

Table S1. XPS Data Analyses of the C 1s and O 1s Spectra of Three CDs

CDs	C 1s			O 1s	
	C=C/C-C	C-O	C=O	C=O	C-O
Y-CD	75.2%	17.9%	6.9%	35%	65%
R-CD	80.9%	14.7%	4.4%	24.9%	75.1%
D-CD	80.1%	13.2%	6.7%	33.5%	66.5%

Table S2. Biexponential Fitting Results of PL Decay of Three CDs

Sample	τ_1/ ns	B_1 / %	τ_2/ ns	B_2 / %	τ(avg)/ ns
Y-CDs	1.92	51.00	7.16	49.00	4.49
R-CDs	3.38	41.70	9.53	58.30	6.97
D-CDs	2.42	32.97	7.15	67.03	5.46

Table S3. Comparison of performances of CDs lasers

Reference	Type of lasers	threshold	lasing wavelength	Working temperature
This work	F-P lasing	33 kW/cm^2	612 nm	250 ºC
[1]	F-P lasing	60 kW/cm^2	561 nm	Room temperature
[2]	F-P lasing	80 kW/cm^2	458 nm	Room temperature

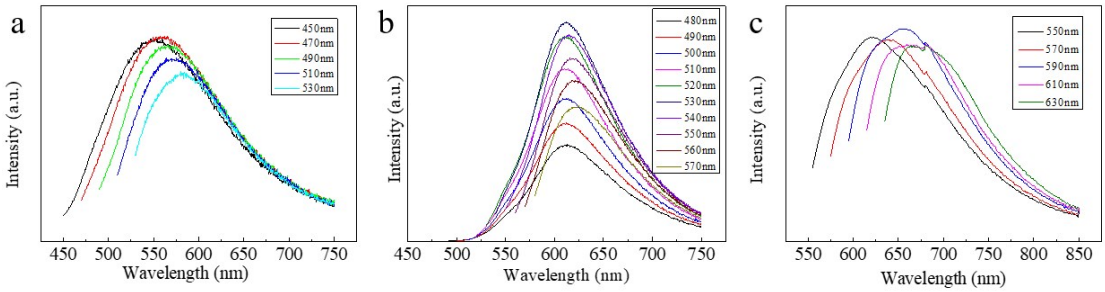

Figure S1. (a), (b), (c) PL emission spectra of Y/R/D-CDs under excitation of different wavelengths of light.

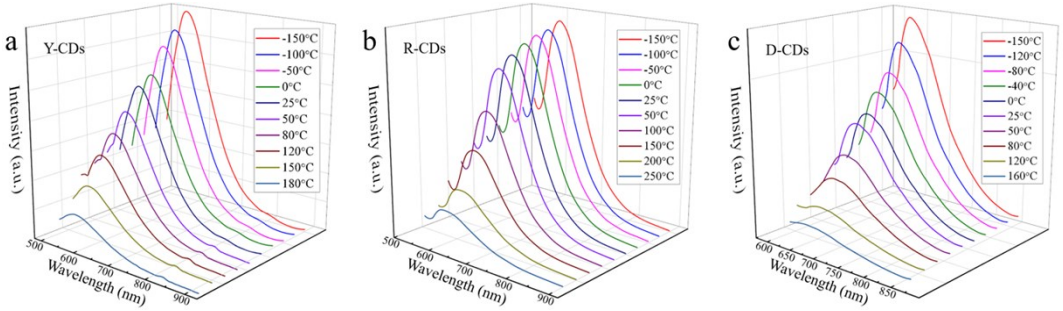

Figure S2. (a), (b), (c) PL emission spectra of Y/R/D-CDs/epoxy composite at different temperatures respectively.

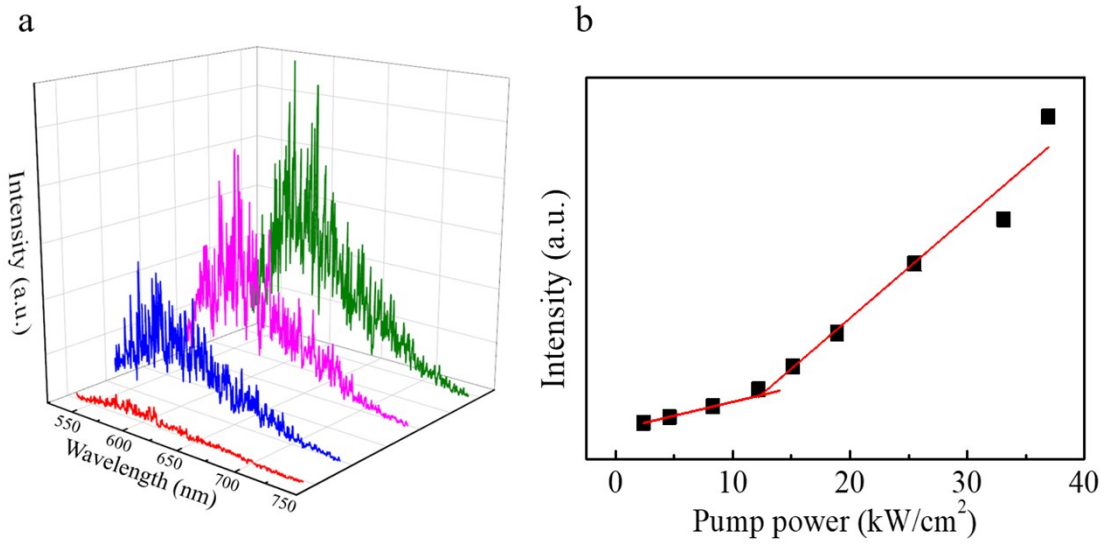

Figure S3. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=22 μm) under different power of excitation light (532 nm) at 50 ºC.

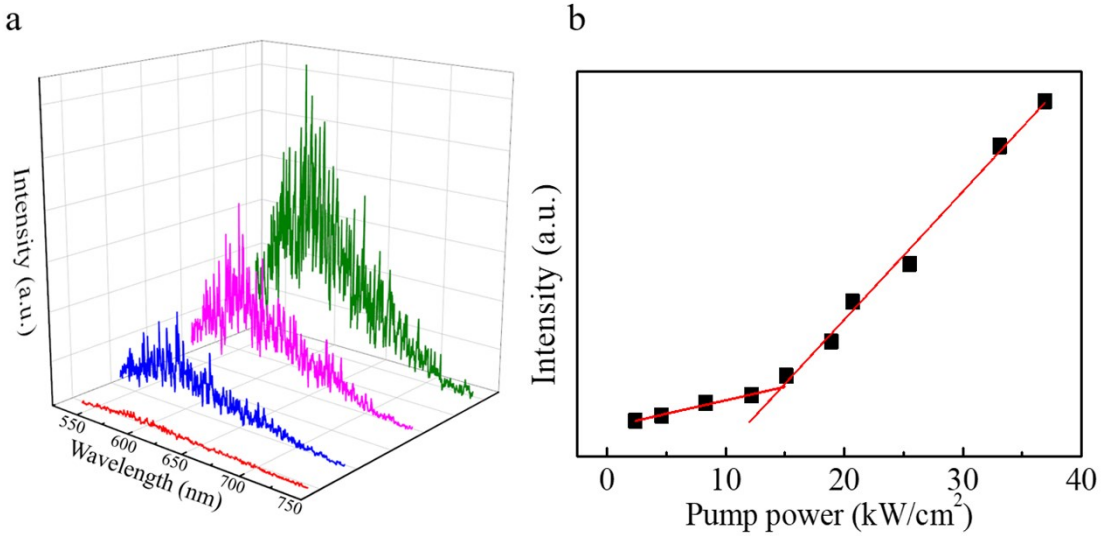

Figure S4. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=22 μm) under different power of excitation light (532 nm) at 80 ºC.

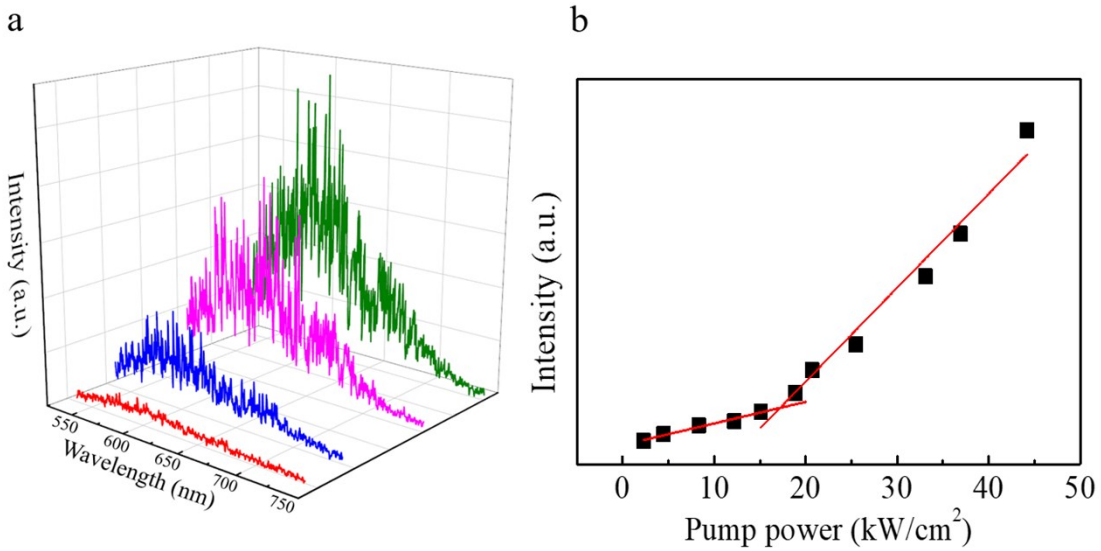

Figure S5. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=22 μm) under different power of excitation light (532 nm) at 120 ºC.

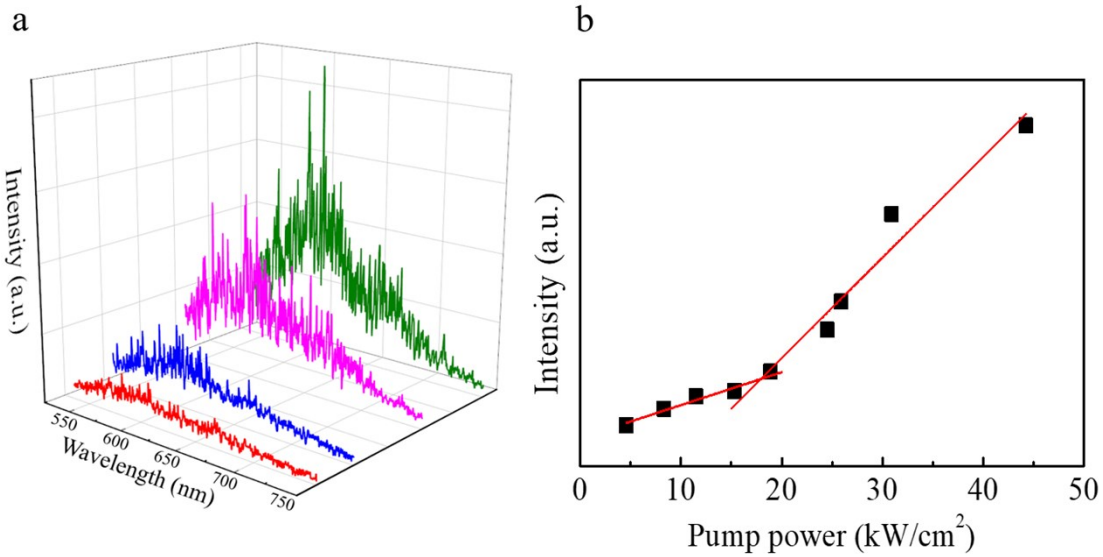

Figure S6. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=22 μm) under different power of excitation light (532 nm) at 160 ºC.

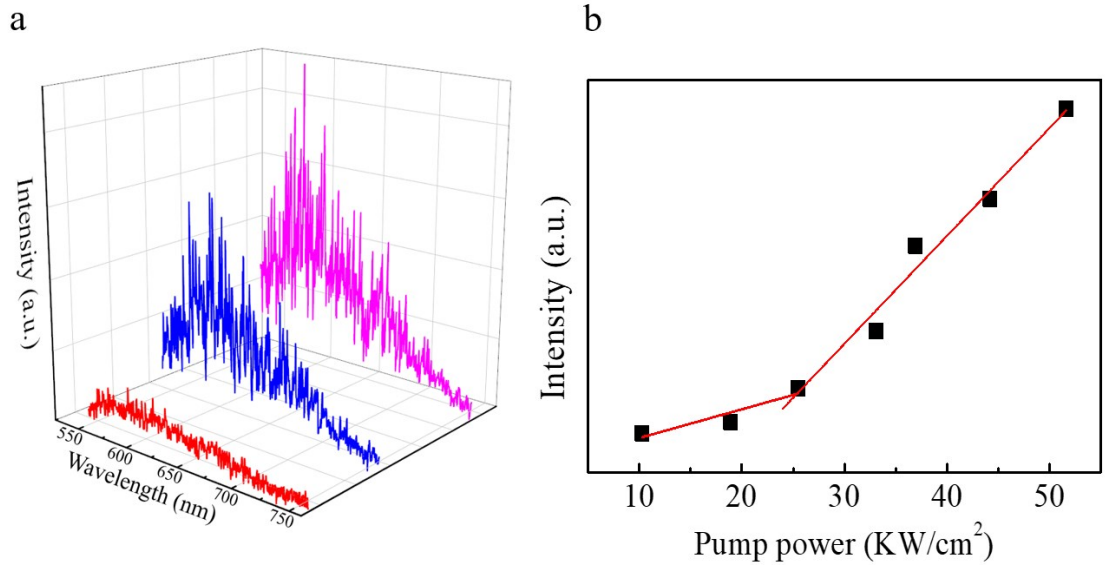

Figure S7. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=22 μm) under different power of excitation light (532 nm) at 200 ºC.

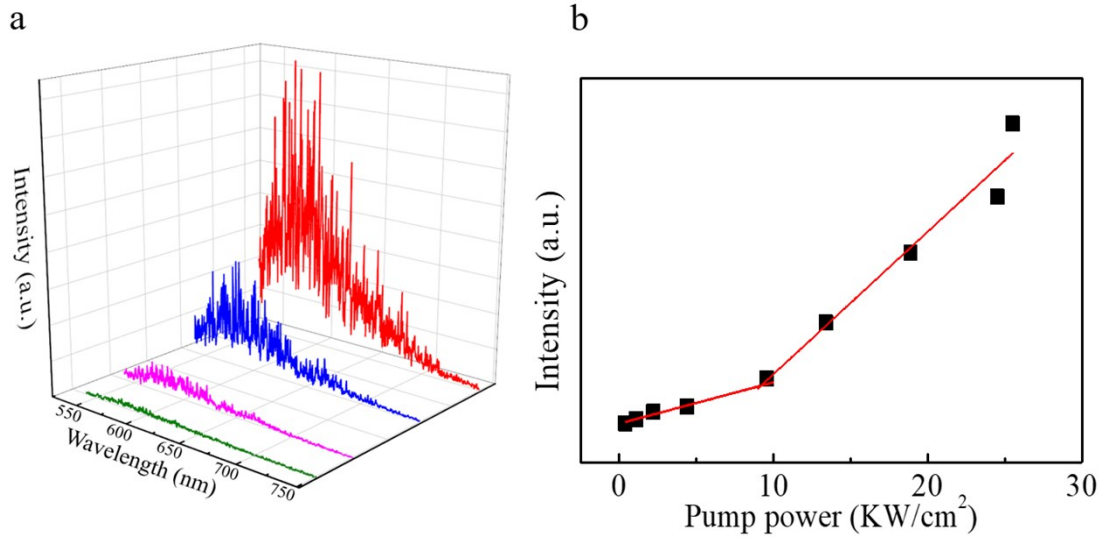

Figure S8. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=15 μm) under different power of excitation light (532 nm) at room temperature.

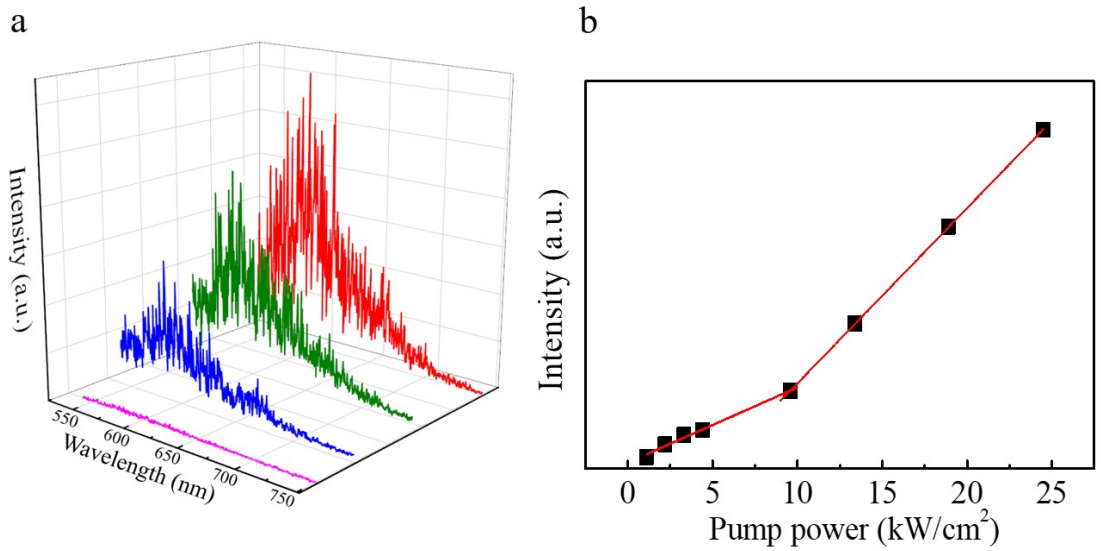

Figure S9. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=15 μm) under different power of excitation light (532 nm) at 50 ℃.

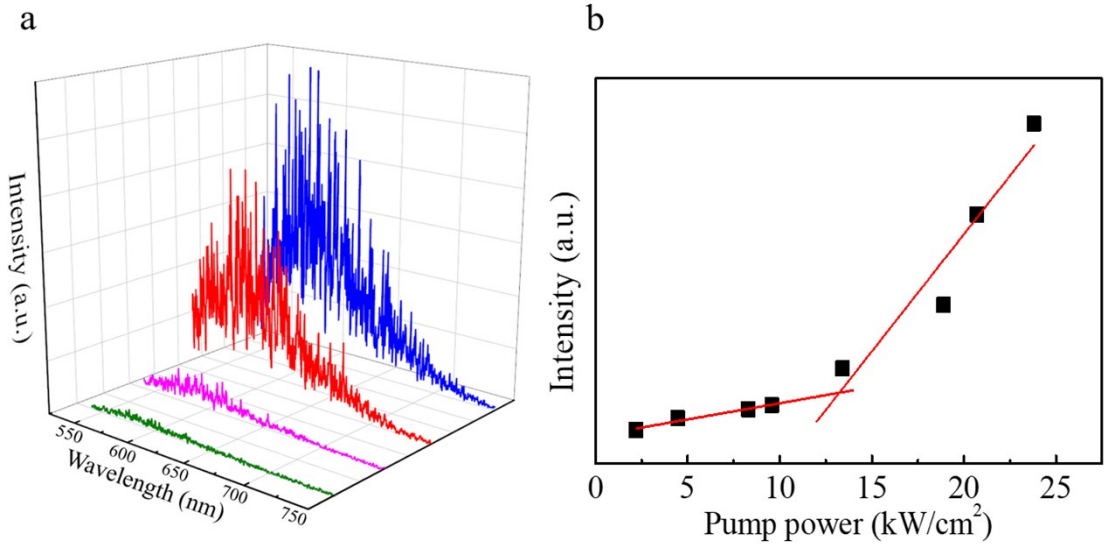

Figure S10. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=15 μm) under different power of excitation light (532 nm) at 100 ºC.

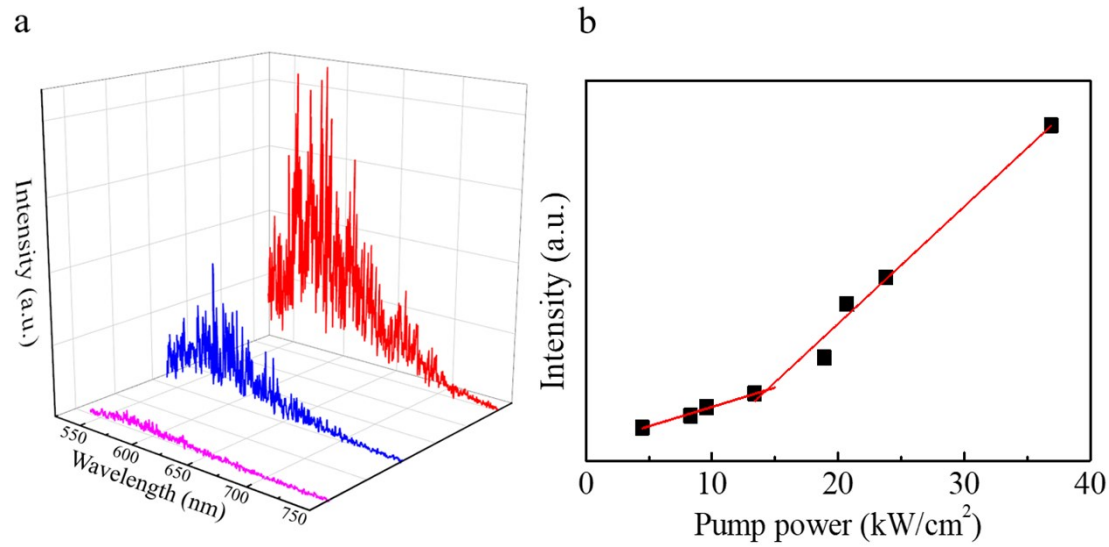

Figure S11. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=15 μm) under different power of excitation light (532 nm) at 150 ºC.

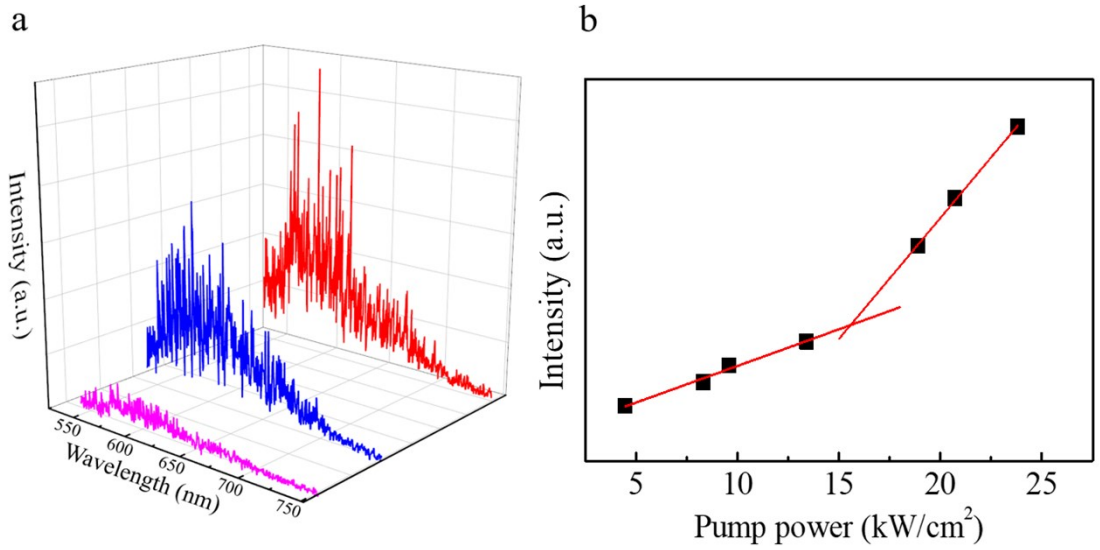

Figure S12. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=15 μm) under different power of excitation light (532 nm) at 200 ºC.

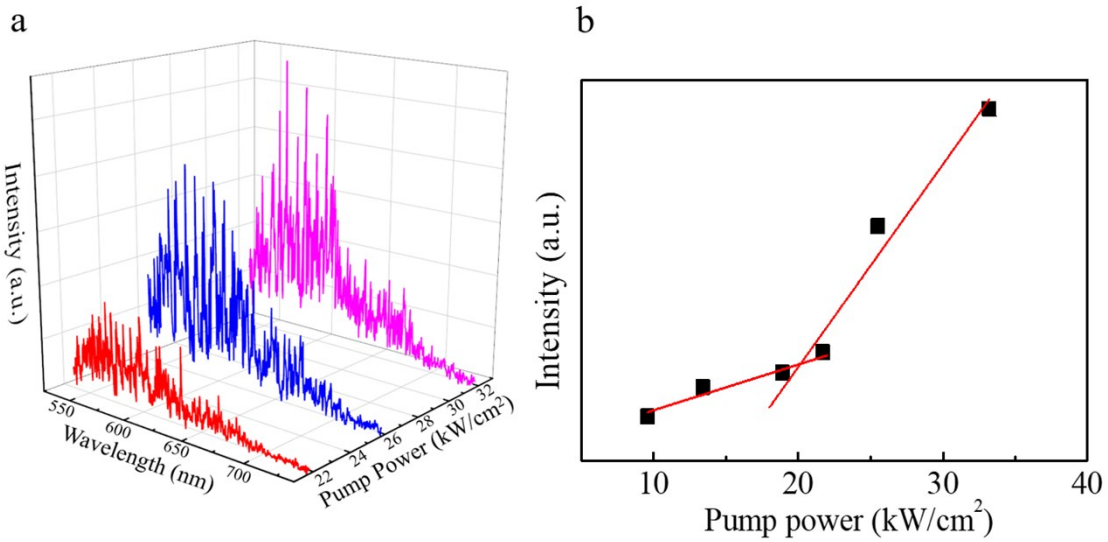

Figure S13. (a), (b) F-P lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=15 μm) under different power of excitation light (532 nm) at 250 ºC.

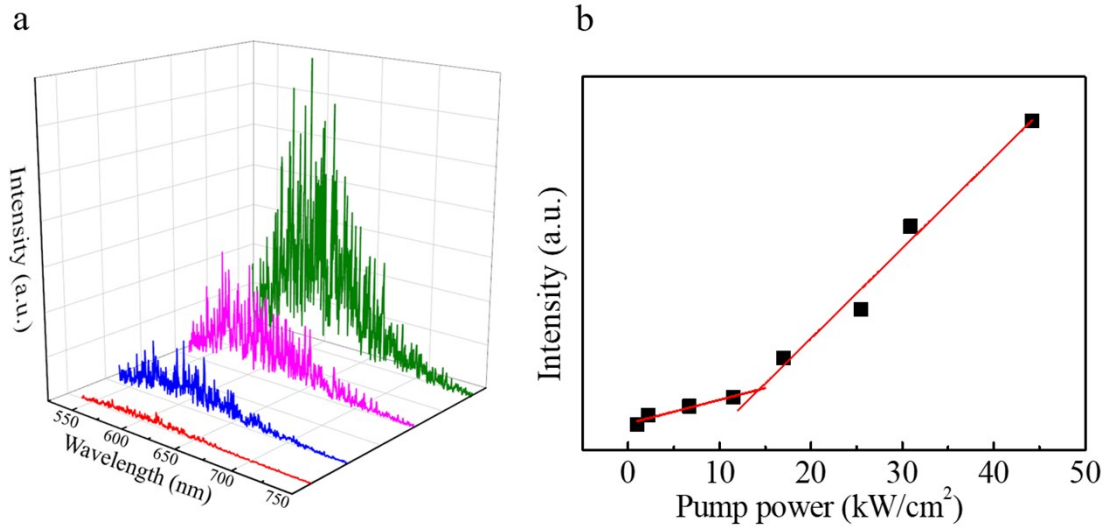

Figure S14. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=27 μm) under different power of excitation light (532 nm) at 50 ºC.

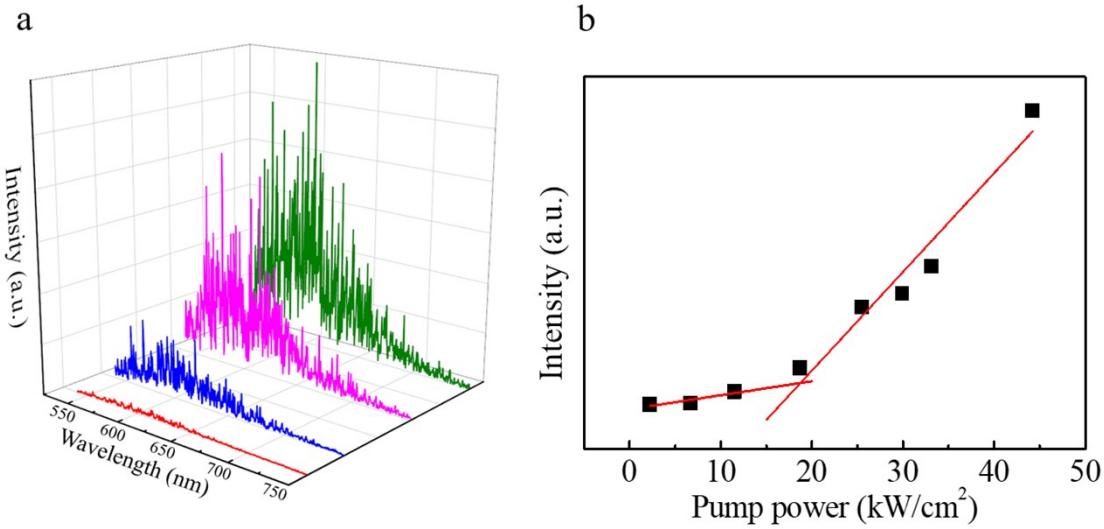

Figure S15. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=27 μm) under different power of excitation light (532 nm) at 80 ºC.

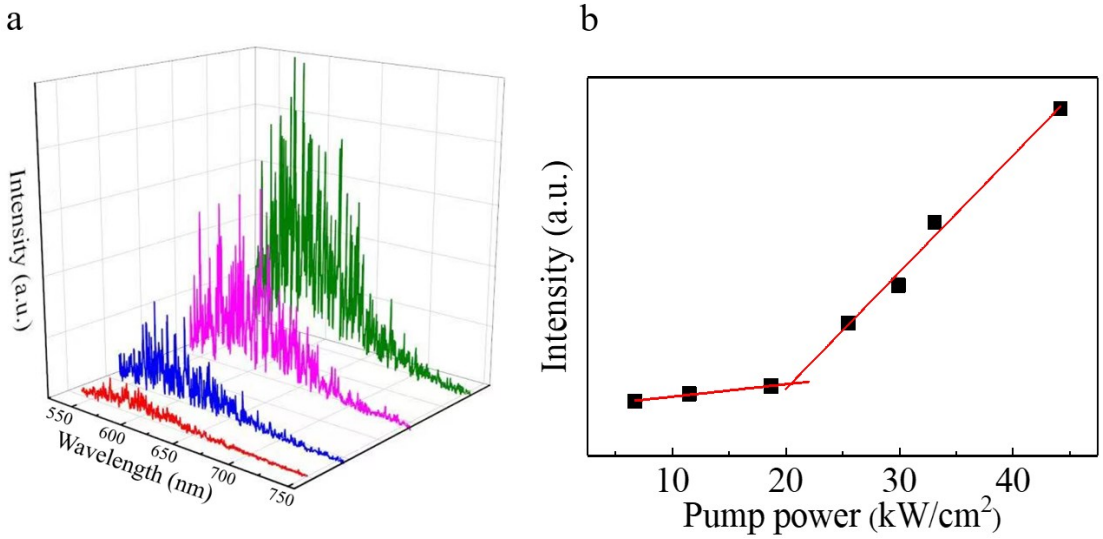

Figure S16. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=27 μm) under different power of excitation light (532 nm) at 120 ºC.

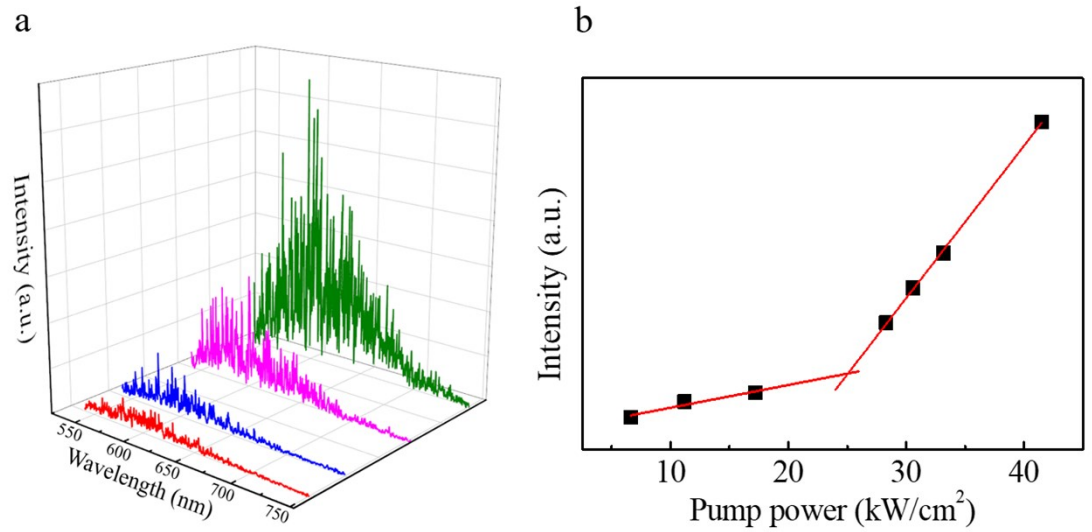

Figure S17. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=27 μm) under different power of excitation light (532 nm) at 160 ºC.

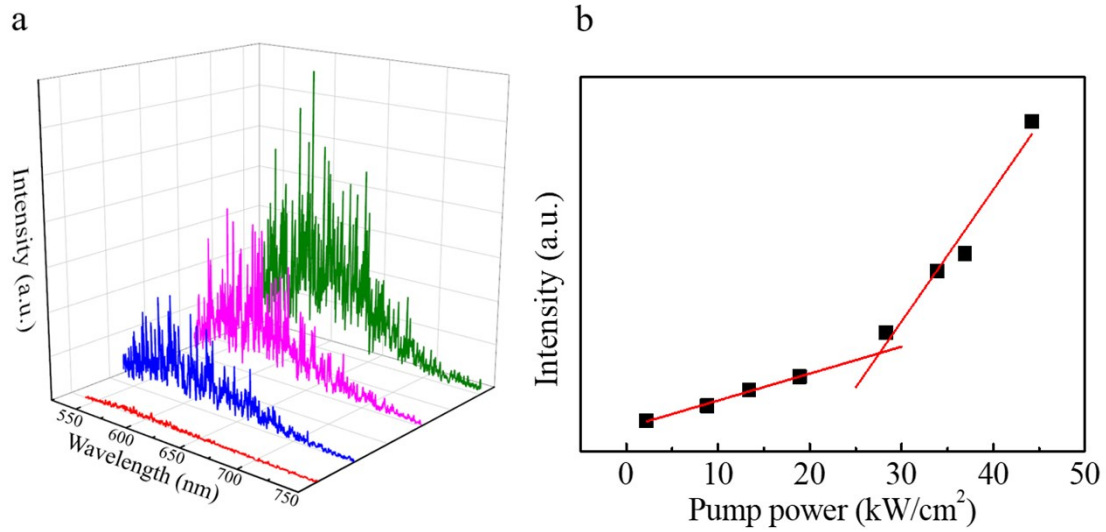

Figure S18. (a), (b) Random lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=27 μm) under different power of excitation light (532 nm) at 200 ºC.

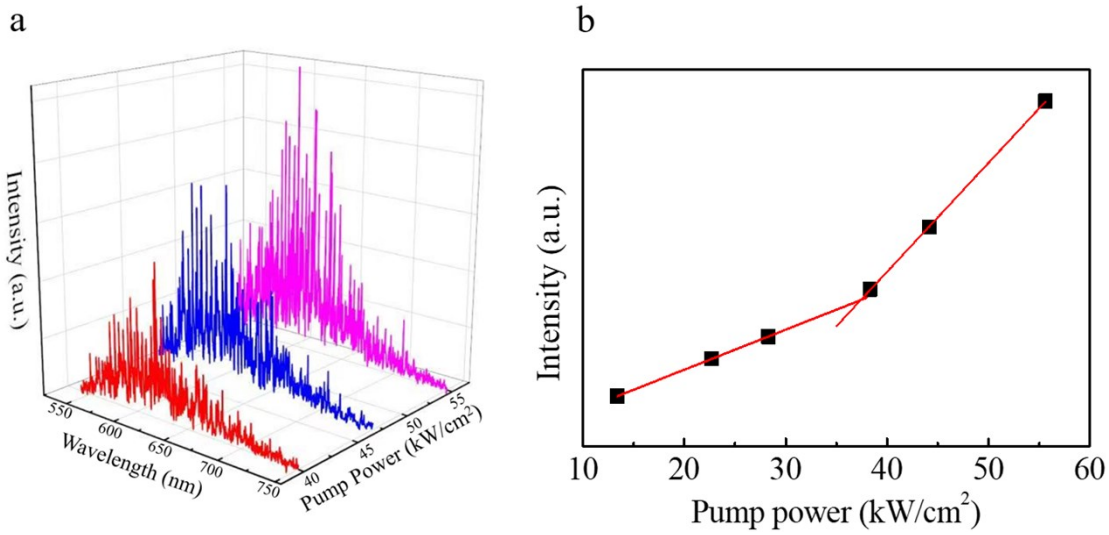

Figure S19. (a), (b) F-P lasing spectra and input excitation power versus output intensity plot of a typical microcavity (L=27 μm) under different power of excitation light (532 nm) at 250 ºC.

[1] S.N. Qu, X.Y. Liu, X.Y. Guo, M.H. Chu, L.G. Zhang, D.Z. Shen, Amplified Spontaneous Green Emission and Lasing Emission From Carbon Nanoparticles, Adv Funct Mater, 24 (2014) 2689-2695.

[2] Y.Q. Zhang, Y.S. Hu, J. Lin, Y. Fan, Y.T. Li, Y. Lv, X.Y. Liu, Excitation Wavelength Independence: Toward Low-Threshold Amplified Spontaneous Emission from Carbon Nanodots, Acs Applied Materials & Interfaces, 8 (2016) 25454-25460.