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Supplementary Document

Wide-bandwidth lasing from C-dots/epoxy nano-composite Fabry Perot cavities with ultralow threshold

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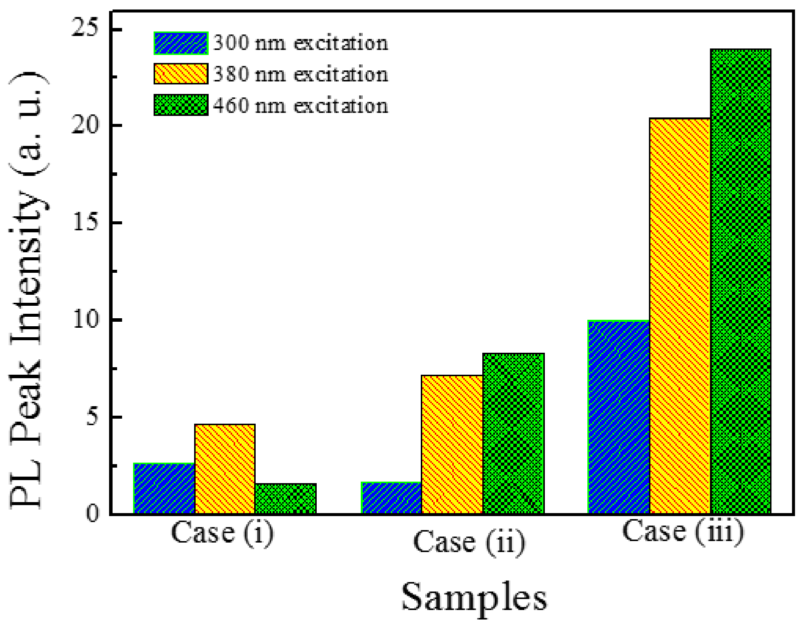

Figure S1 PL peak intensity of the C-dots fabricated by using case (i) 1:0, case (ii) 1:10 and case (iii) 1:20 of citric acid:A-2120 mass ratio. The C-dots were under excitation by 300, 380 and 450 nm lasing light. In case (i), the corresponding emission intensity is the lowest for all the excitation wavelengths. For case (iii), the corresponding emission intensity is the highest for all the excitation wavelengths. For case (ii), the magnitude of emission intensity is in between cases (i) and (iii) due to the co-existence of similar amount of COOH and organosiloxane functional groups.

Justification to relate optical gain with the pump threshold and the role of organosiloxane functional groups

We repeated our studies on the lasing characteristics of different type of the C-dots gain materials by using identical configuration of Fabry Perot cavity (same as that described in the manuscript) and excited at their optimal excitation wavelength. The Fabry Perot cavity consists of a capillary tube of length 2 mm and inner diameter of 100 μm to hold the C-dots gain medium, and two mirrors (one is an Al mirror and the other one is a DBR mirror) to provide optimized optical feedback at the operation wavelength. In this experiment, it is reasonable to assume that the only different is the use of C-dots materials. The threshold gain, g_{th}, of the Fabry Perot cavity can be given as [R1]

$$g_{th} = \alpha_i + \frac{1}{2L}\ln\left(\frac{1}{R_1R_2}\right) \tag{S1}$$

where α_i is the waveguide and material losses, L is the cavity length, R_1 and R_2 are the reflectivity of the two mirrors of the Fabry Perot cavity. As all of the C-dots materials have similar transparency near their operation wavelength, we can assume α_i is mainly waveguide dependent. Hence, the minimum optical gain required to excite the Fabry Perot is equal to g_{th} as defined by (S1).

As g_{th} is mainly dependent on the configuration of laser cavities (i.e., independent on the optical properties of C-dots), we may able to use the pump threshold to investigate the optical quality of the luminescent C-dots materials. This is because the pump threshold, P_{th}, of the Fabry Perot lasers can be written as [R1]

$$P_{th} \approx \frac{D}{v_g \alpha_{ab} \tau_N}\left(\frac{g_{th}}{a_N} + N_0\right) \approx \underbrace{\frac{Dg_{th}}{v_g}}_{constant} \times \underbrace{\frac{1}{\alpha_{ab}}}_{\substack{absorption\\ of\\ C\text{-}dots}} \times \underbrace{\frac{1}{a_N \tau_N}}_{\substack{gain\\ related\\ parameters}} \tag{S2}$$

where D is the diameter of the capillary tube, v_g is the group velocity, α_{ab} is the absorption coefficient at the pump wavelength and τ_N is the non-radiative lifetime. a_N is the linear gain coefficient and N_0 is the carrier concentration at transparent of the C-dots materials. In the derivation of (S2), we have assumed that the optical gain of C-dots is linear proportional to the carrier concentration and the value of N_0 is smaller than its threshold value in order to simplify the analysis.

As we can see from (S2), the optical characteristics of C-dots are mainly related to P_{th} (i.e., α_{ab}, a_N and τ_N are mainly dependent on the optical properties of C-dots). Hence, it is reasonable to compare the performance of C-dots by using P_{th}.

Table S1 shows the measured values of α_{ab}, P_{th} and quantum yield (QY) of different types of C-dots materials. The values of α_{ab} for different C-dots materials were found to have similar values at their excitation wavelength. Hence, the significant reduction of P_{th} (reduced by 2 order of magnitude) for the C-dots materials reported in this manuscript represents the huge increase of $a_n\tau_n$. Furthermore, QY is defined as the ratio of the number of photons emitted to the number of photons absorbed so that we can express QY as [R2]

$$QY = \frac{total\ no\ of\ \ photons\ emitted}{total\ no\ of\ \ photons\ absorbed} = \frac{\tau_N}{\tau_N + \tau_R} \quad \text{(S3)}$$

where τ_R and τ_N are the radiative and nonradiative lifetime respectively. The increase of QY may be due to the increase of τ_N or the decrease of τ_R or simultaneously increase and decrease of τ_N and τ_R, respectively. For the situation that the increase of QY is due to the increase of τ_N, the increase of τ_N is still less than an order of magnitude. Hence, it is still reasonable to attribute the reduction of P_{th} is related to the increase of a_N or optical gain. This is because the rate of increase of QY is much lower than that of the value of P_{th}. Therefore, it is reasonable to draw the conclusion that the reduction of P_{th} implies the increase of optical gain of the C-dots. In addition, the increase of optical gain is in order of magnitude.

It is also interested to know whether the C-dots given in Table S1 have organosiloxane functional groups. Figure S2 plots of FTIR spectra of the C-dots. It is noted that C-dots reported previously have lesser organosiloxane functional groups due to the co-existence of surface functional -OH groups. In addition, the presence of organosiloxane functional groups can improve the PL emission intensity even for the C-dots fabricated by different method.

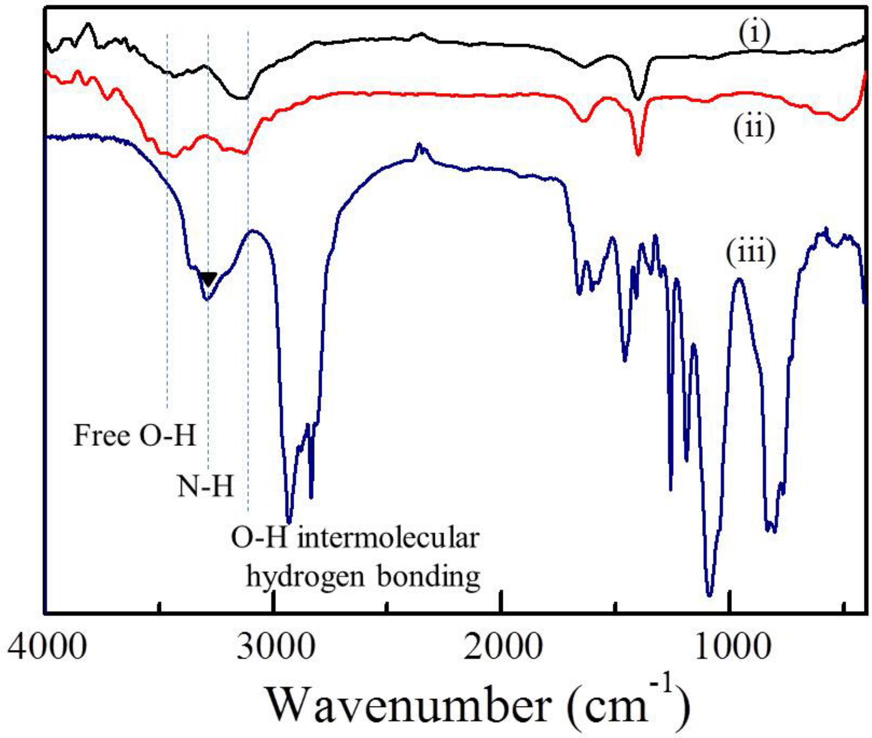

Figure S2 FTIR spectra of (i) C-dots [R3] with -OH and -COOH functional groups, (ii) graphene quantum dots with -OH and ester functional groups, and (iii) C-dots [this paper] with organosiloxane functional groups. It is noted that only C-dots [this paper] have no -OH and -COOH functional groups.

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Table S1 Comparison of lasing performance between C-dots in [R3], [R4] and that given in this manuscript with citric acid: A-2120 ratio of 1:20 and concentration of 50%

Type	P_{th} (kW/cm^2)	α_{ab} (/cm)	QY (%)	Excitation wavelength	Operation wavelength
C-dots [R3]	204	16.9	10	266 nm	~380 nm
graphene quantum dots [R4]	81	18.7	16	266 nm	~400 nm
C-dots [50%, this paper]	0.2	24.1	68	450 nm	~540 nm

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