N-doped carbon dot with surface dominant non-linear optical property

(Supporting Information)

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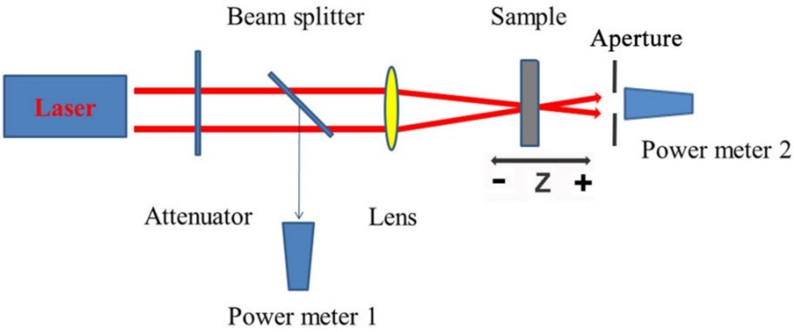

Figure S1. Experimental setup for Z-scan measurement.

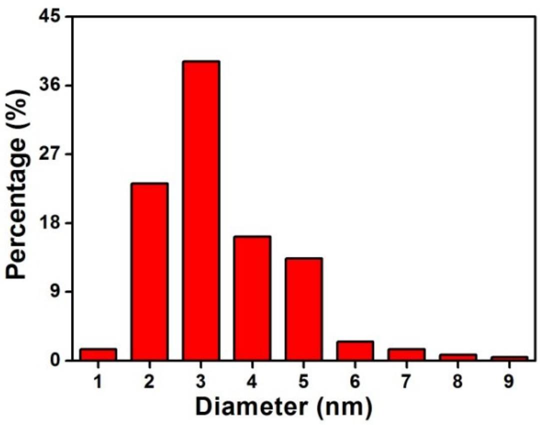

Figure S2. The particle sizes histogram of N-CDs.

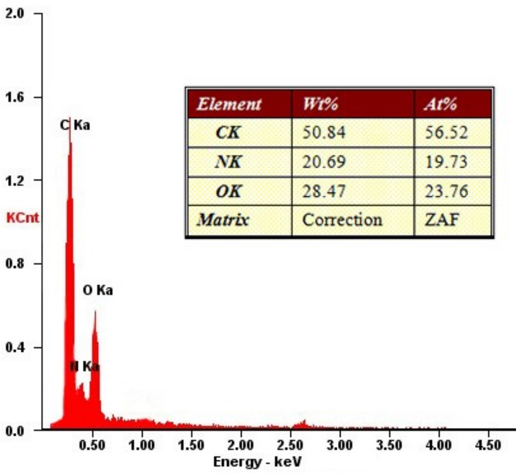

Figure S3. Energy dispersive x-ray spectroscopy analysis with element relative content of N-CDs in the inset.

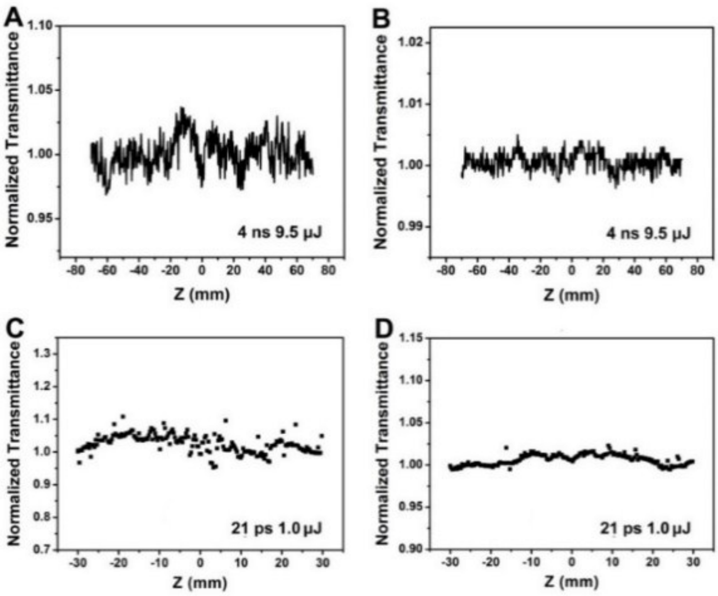

Figure S4. Close aperture (A and C) and open aperture (B and D) Z-scans of water measured under 532 nm, 4 ns (A and B) and 21 ps (C and D) laser excitation.

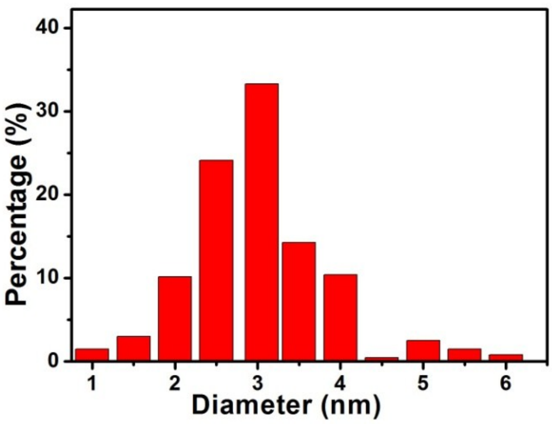

Figure S5. The particle sizes histogram of RN-CDs.

The Z-scan is a simple and popular experimental technique to measure the intensity dependent third-order nonlinear susceptibility of the materials. It allows the simultaneous measurement of both the nonlinear refractive index γ and the nonlinear absorption coefficient β. From these parameters, the real and the imaginary components of the third-order nonlinear susceptibility $\chi^{(3)}$ can be calculated using the following relations [26]:

$$T(Z) = \frac{\alpha_0}{\sqrt{\pi}\beta I_i(Z)(1 - e^{-\alpha_0 L})}\int_{-\infty}^{\infty} ln\left[1 + \beta I_i(Z)\frac{1 - e^{-\alpha_0 L}}{\alpha_0}e^{-\tau^2}\right]d\tau \qquad (1)$$

$$\gamma = \frac{\Delta T_{p-v}\lambda}{0.812\pi I_0(1-S)^{0.25}L_{eff}} \qquad (2)$$

$$\chi_{Re}^{(3)}(esu) = \frac{cn_0^2\gamma}{120\pi^2} \quad (3)$$

$$\chi_{Im}^{(3)}(esu) = \frac{n_0^2c^2\beta}{240\pi^2\omega} \quad (4)$$

$$\chi^{(3)} = \left[\left(\chi_{Im}^{(3)}\right)^2 + \left(\chi_{Re}^{(3)}\right)^2\right]^{\frac{1}{2}} \quad (5)$$

where L_{eff} is the effective sample thickness, α_0 is the linear absorption coefficient at the excitation wavelength λ, L is the sample length, I_0 is the laser peak irradiance on the focal plane, z_0 is the Rayleigh length, z is the position of the sample, $\Delta T_{p\text{-}v}$ is the difference between the peak and the valley of the normalized transmittance, and S is the linear transmittance of the aperture, defined as $S = 1 - \exp(-2r_a^2/w_a^2)$, with r_a being the radius of the aperture and w_a being the beam radius at the aperture.

Table S1. The third-order nonlinear optical parameters of different kinds of CDs under 532 nm laser excitation.

Sample	Laser	γ (10^{-17} $m^2 \cdot W^{-1}$)	β (10^{-11} $m \cdot W^{-1}$)	$\chi^{(3)}$ (10^{-12} esu)	$\chi^{(3)}/C^a$ (10^{-12} $esu \cdot L \cdot g^{-1}$)	References
N-CDs	4 ns	2.73	12.7	12.5	19.8	This work
N-CDs	21ps	-0.161	0.196	0.725	1.15	This work
B-CDs[b]	4 ns	-0.4	5.1	0.50	0.86	29
B-CDs	35 ps	-0.0034	-	0.0029	0.005	29
CDs[c]	35 ps	-0.043	-	0.048	0.026	30

[a)]C: sample concentration; [b)]B-CDs: boron doped carbon dots; [c)]CDs: amorphous carbon dots.