Supplementary information

Investigation of broadband optical nonlinear absorption and transient nonlinear refraction in a fluorenone-based compound

Xingzhi Wu [a,]*, Jinchong Xiao [b], *Yanbing Han* [c], *Jiabei Xu* [a], *Wenfa Zhou* [c], *Yang Li* [a],, *Yu Fang* [a], *Yongqiang Chen* [a], *Quanying Wu* [a] *and Yinglin Song* [c, d,]*

[a] Jiangsu Key Laboratory of Micro and Nano Heat Fluid Flow Technology and Energy Application, School of Physical Science and Technology, Suzhou University of Science and Technology, Suzhou 215009, China

[b] College of Chemistry and Environment Science, Key Laboratory of Chemical Biology of Hebei Province, Hebei University, Baoding 071002, China

[c] Department of Physics, Harbin institute of technology, Harbin 150001, China

[d] Department of Physics, Soochow University, Suzhou 215123, China

Corresponding Author

*E-mail: wuxingzhi@usts.edu.cn

* E-mail: ylsong@hit.edu.cn

1. Characterization

Figure S1 ^{1}H NMR

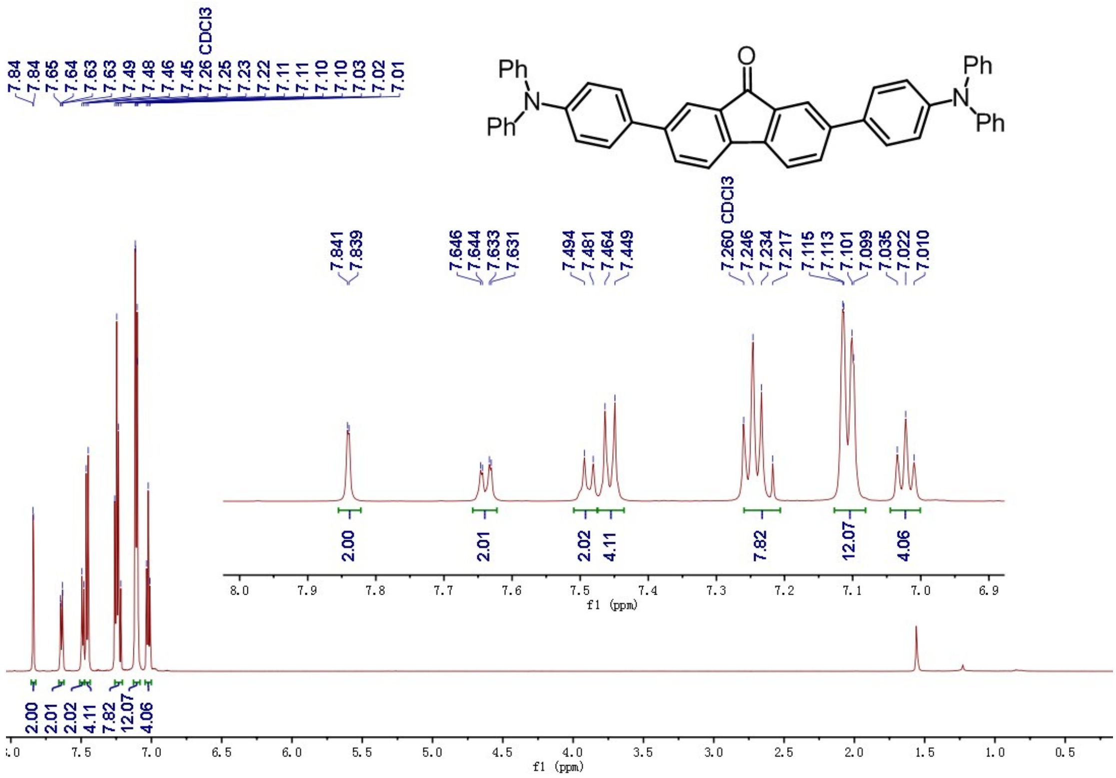


Figure S2 ^{13}C NMR

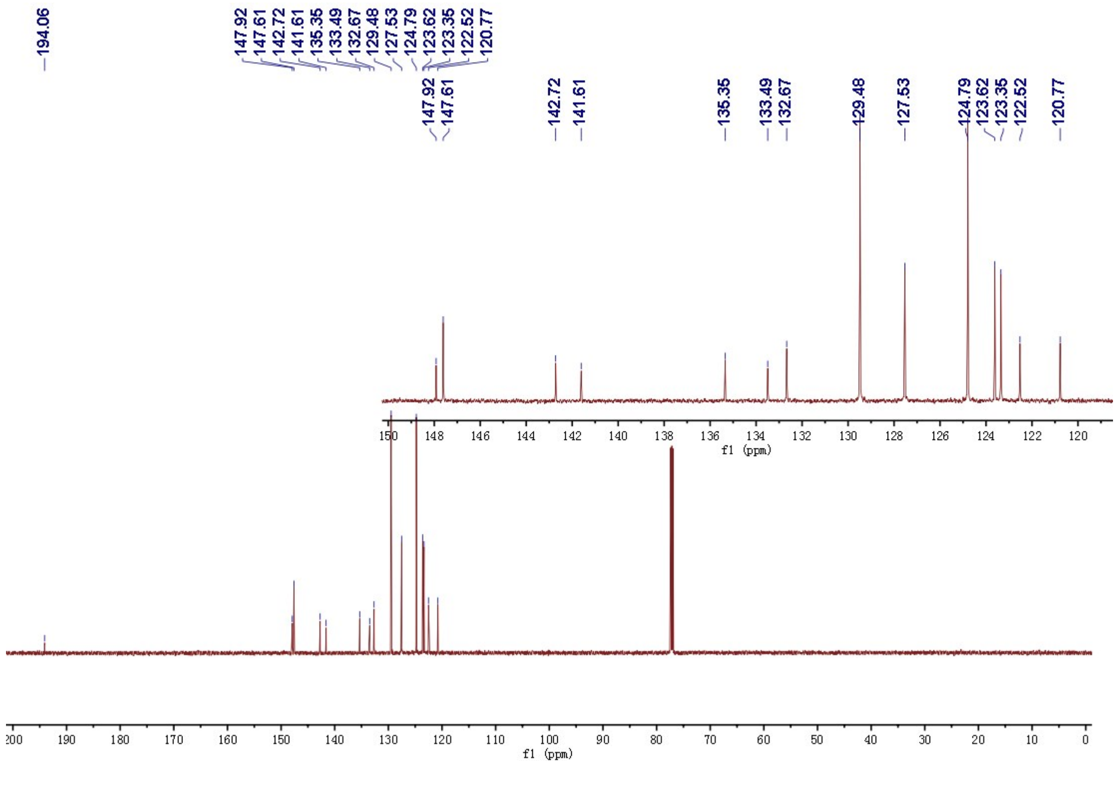
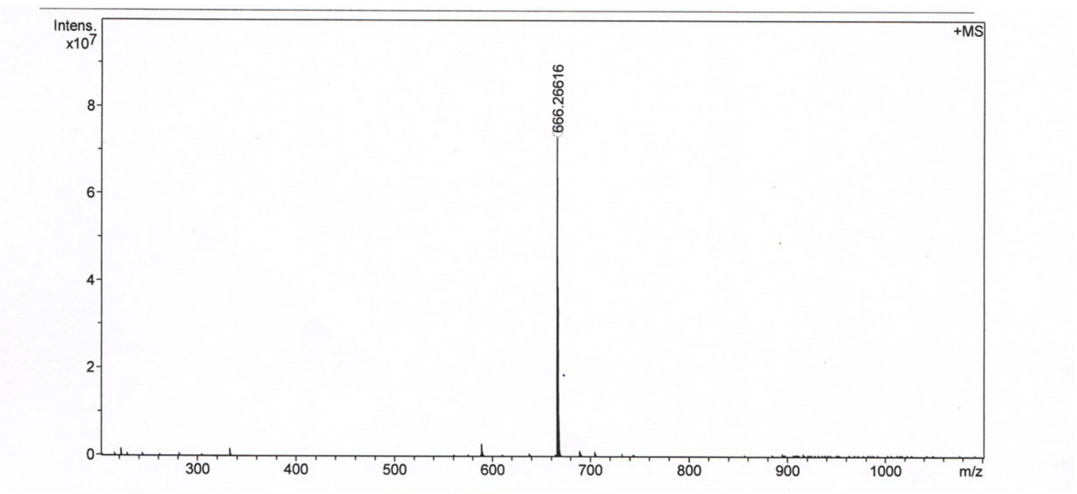
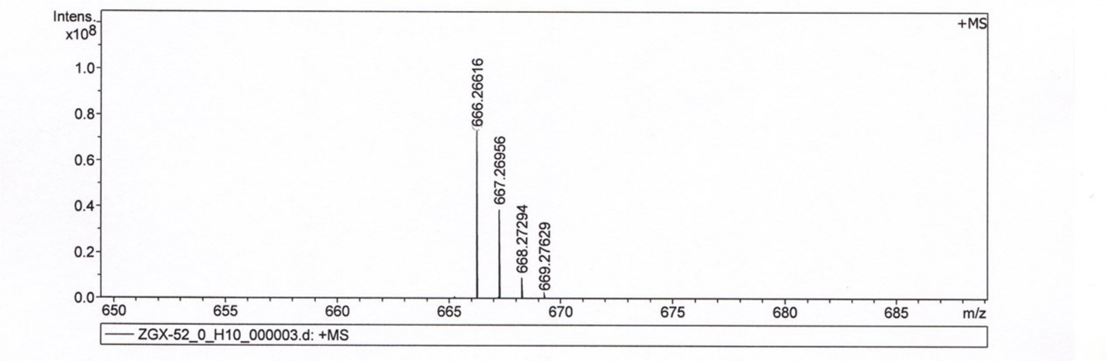


Figure S3 MS

MALDI-TOF



HR-MS



2. Transient absorption spectrum

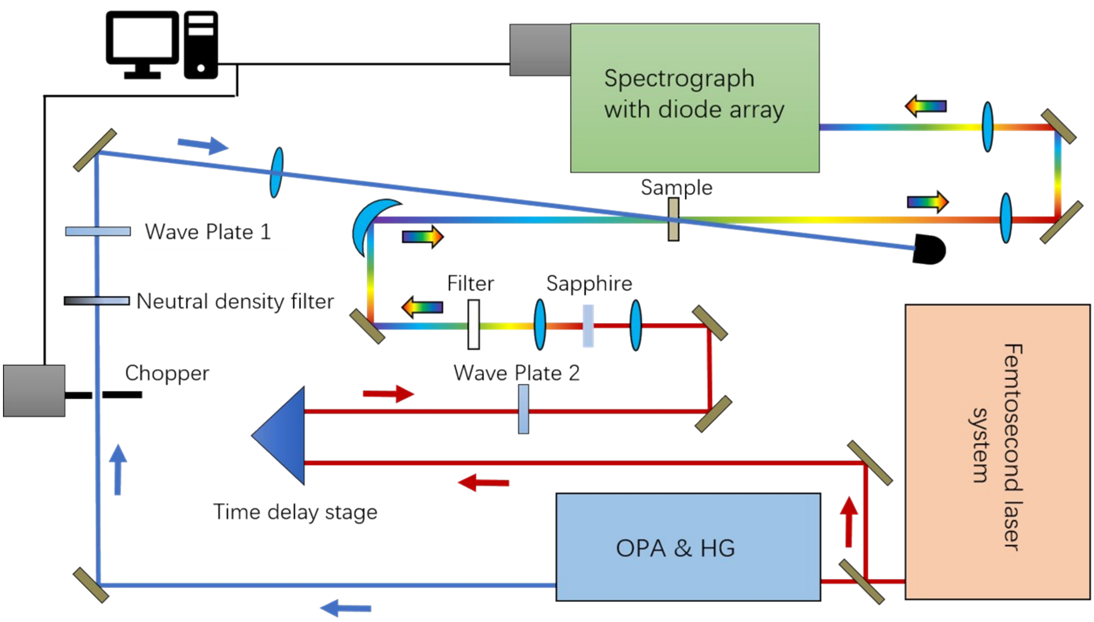


Figure S4 The experimental setup of TAS

3. Global and Target analysis of TAS

The total TAS ($\psi(t, \lambda)$) could be numerically reconstruct with several components, which could be express as:

$$\psi(t,\lambda)=\sum_n c_n(t)\times\varepsilon_n(\lambda)$$

$c_n(t)$ and $\varepsilon_n(\lambda)$ represent the concentration of time and the featured spectrum of each component n. As a result, $c_n(t)$ could be expressed as lifetime τ in single exponential decay $c_n = \exp(-t/\tau_n)$ and the corresponding spectrum of each component could be extracted. The IRF here is given by the convolution of pump and probe pulses, which was estimated to be 0.2 ps. The details of TAS and global & target analysis could be found in Refs listed below.

[1] I.H.M. van Stokkum, D.S. Larsen, R. van Grondelle, Global and target analysis of time-resolved spectra, Bba-Bioenergetics 1657(2-3) (2004) 82-104.

[2] C. Ruckebusch, M. Sliwa, P. Pernot, A. de Juan, R. Tauler, Comprehensive data analysis of femtosecond transient absorption spectra: A review, Journal of Photochemistry and Photobiology C: Photochemistry Reviews 13(1) (2012) 1-27.

[3] R. Berera, R. van Grondelle, J.T.M. Kennis, Ultrafast transient absorption spectroscopy: principles and application to photosynthetic systems, Photosynth Res 101(2-3) (2009) 105-118.

4. Rate equations in POPP simulation

Simplified rate equations are built as follows:

$$\frac{dN_{S_0}}{dt}=-\frac{\sigma_{e,S_0} I_e N_{S_0}}{\hbar\omega}-\frac{\beta I_e^2}{2\hbar\omega}+\frac{N_{S_1}}{\tau_{S_1}}$$

$$\frac{dN_{S_3}}{dt}=\frac{\sigma_{e,S_0} I_e N_{S_0}}{\hbar\omega}+\frac{\beta I_e^2}{2\hbar\omega}-\frac{N_{S_3}}{\tau_{S_3}}$$

$$\frac{dN_{S_2}}{dt}=\frac{N_{S_3}}{\tau_{S_3}}-\frac{N_{S_2}}{\tau_{S_2}}$$

$$\frac{dN_{S_1}}{dt}=\frac{N_{S_2}}{\tau_{S_2}}-\frac{N_{S_1}}{\tau_{S_1}}$$

With N_{Sn} represents the population density of effective energy state S_n. $\sigma_{e,Sn}$ and $\sigma_{p,Sn}$ are absorptive cross section of S_n for pump beam and probe beam, β is the two-photon absorption coefficient. I_e and I_p stand for pump intensity and probe intensity. τ_n represents lifetimes of each effective state. We assume that electrons are only pumped from ground state to the LE state S_2 during the pulse time. After the effect of pump pulse, excited state electrons decay to S_1 and finally relax to ground state S_0.

And the modulation on intensity (I_p) and phase (φ_p) of the probe beam can be represented as:

$$\frac{dI_p}{dz} = -\left(\sum_{n\geq 1}\sigma_{p,S_n} N_{S_n} + 2\beta I_e\right) I_p$$

$$\frac{d\varphi_p}{dz} = k\left(\sum_{n\geq 1}\Delta\eta_{p,S_n} N_{S_n} + 2n_2 I_e\right)$$