Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary Information

Effect of Intramolecular Charge Transfer Processes on Amplified Spontaneous Emission of D-π-A Aggregation-Enhanced Emission Molecules

Yuanzhao Li[1], Pengbo Han[1], Xiaowei Zhang[1], Jiadong Zhou[1], Xianfeng Qiao[1], Dezhi Yang[1], Anjun Qin[1], Ben Zhong Tang[1,2]*, Junbiao Peng[1]* and Dongge Ma[1]**

[1]Center for Aggregation-Induced Emission, Institute of Polymer Optoelectronic Materials and Devices, Guangdong Provincial Key Laboratory of Luminescence from Molecular Aggregates, Guangdong-Hong Kong-Macao Joint Laboratory of Optoelectronic and Magnetic Functional Materials, State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou 510640, People's Republic of China,
E-mail: msdgma@scut.edu.cn, msqinaj@scut.edu.cn, psjbpeng@scut.edu.cn

Prof. B. Z. Tang
[2]Shenzhen Institute of Molecular Aggregate Science and Engineering, School of Science and Engineering, The Chinese University of Hong Kong, Shenzhen, 2001 Longxiang Boulevard, Longgang District, Shenzhen City, Guangdong 518172, China
E-mail: tangbenz@cuhk.edu.cn

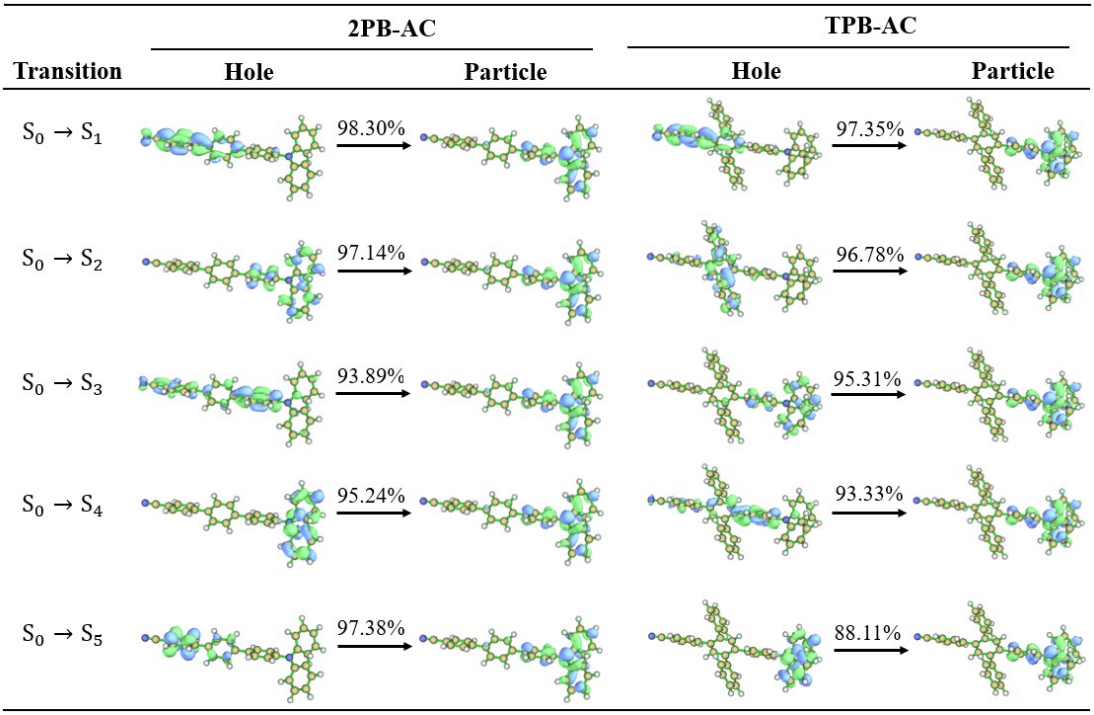

Fig. S1 The calculated NTO distribution of singlet states of 2PB-AC and TPB-AC (Calculation accomplished by Multiwfn 3.3.8. [1]).

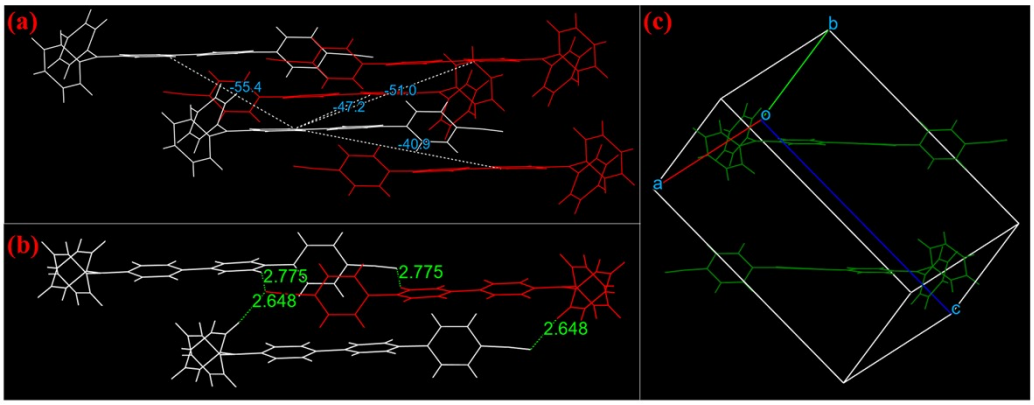

Fig. S2 Single crystal structure of 2PB-AC. (a) First strongest molecular potentials, unit of kJ mol^{-1}. (b) Adjacent molecules distances of CH···N, unit of Å. (c) Unit cell.

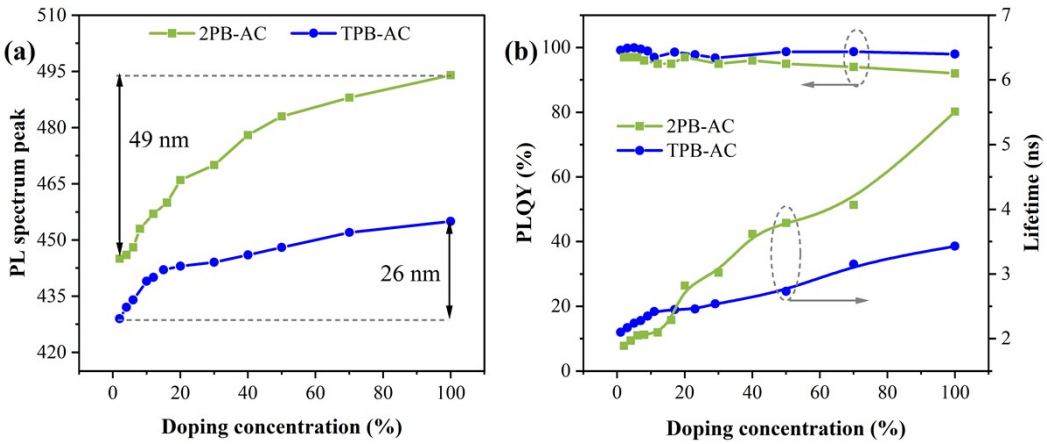

Fig. S3 (a) PL peaks of 2PB-AC and TPB-AC as a function of doping concentration. (b) PL quantum efficiency values and excited state lifetimes of 2PB-AC and TPB-AC as a function of doping concentration.

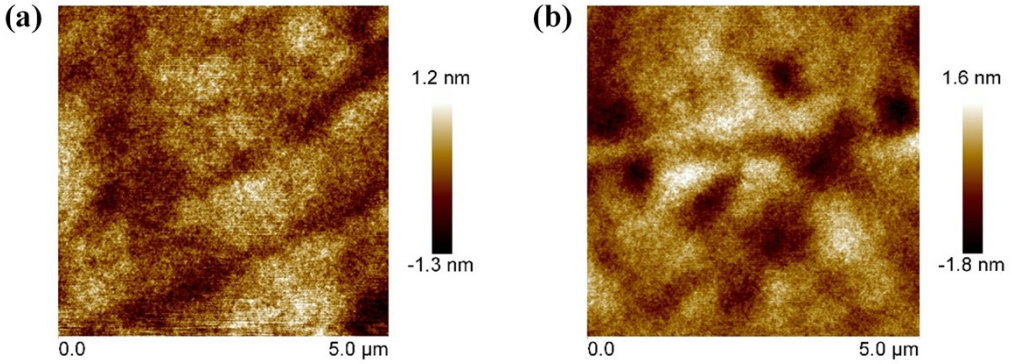

Fig. S4 AFM images of (a) TPB-AC neat film and (b) 2PB-AC neat film.

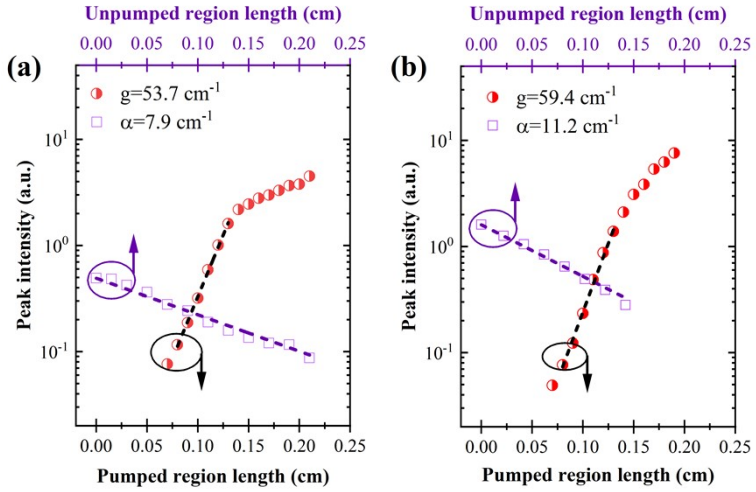

Fig. S5 Loss coefficient (pink color) and net gain coefficient fitting of (a) TPB-AC neat film at the pump energy density of 24 μJ cm^{-2} and (b) CBP:7wt%2PB-AC doped film at the pump energy density of 18 μJ cm^{-2}.

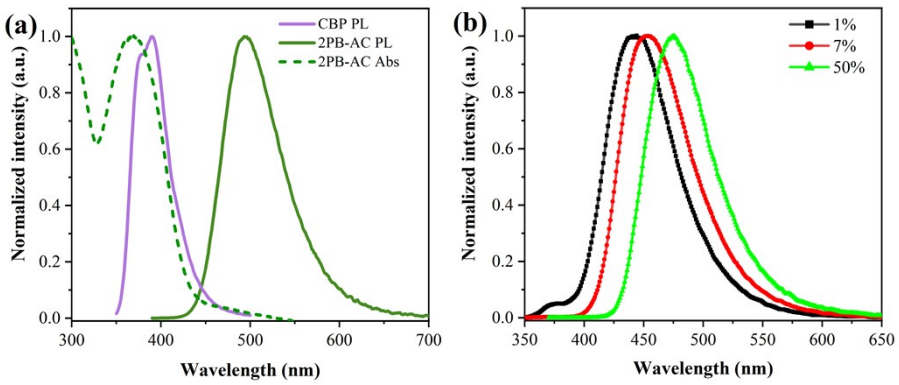

Fig. S6 (a) Normalized absorption spectrum of 2PB-AC neat film (green color) and PL spectra of 2PB-AC (green color) and CBP (pink color) neat films. (b) Normalized PL spectra of CBP:2PB-AC doped films at different concentrations. The excitation wavelength is 350 nm.

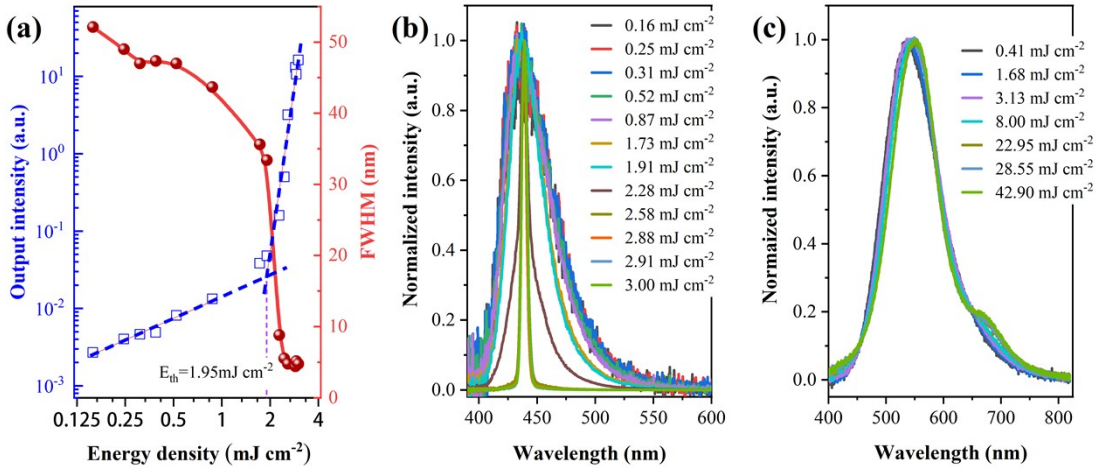

Fig. S7 (a) Output intensity and spectrum FWHM of 2PB-AC in TOL as a function of the pumped energy density. Normalized emission spectra of (b) 2PB-AC in TOL and (c) 2PB-AC in DMF under different pumped energy densities.

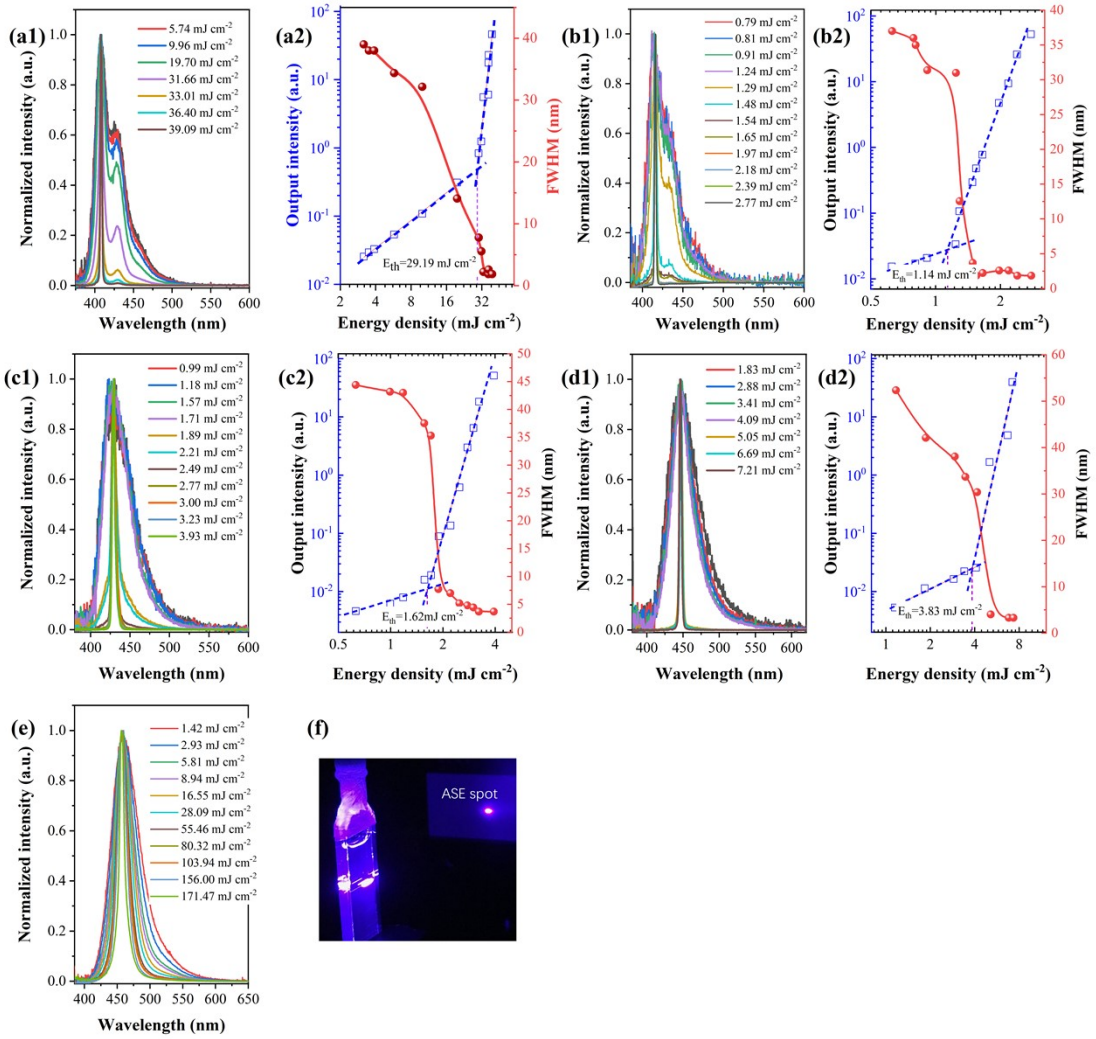

Fig. S8 PL intensity and FWHM of spectra as a function of the pumped energy density and the normalized PL spectra under different pumped energy densities in 2PB-AC mixed solutions (0.25 mg/ml): (a1), (a2) V_{HEX}:V_{TOL} = 10:0 (saturated solution, smaller than 0.25 mg/ml); (b1), (b2) V_{HEX}:V_{TOL} = 8:2; (c1), (c2) V_{HEX}:V_{TOL} = 1:1; (d1), (d2) V_{HEX}:V_{CB} = 1:1; (e) V_{HEX}:V_{CB} = 2:8. (f) Photograph of ASE spot for V_{HEX}:V_{TOL} = 8:2 solution when pumped density exceeding ASE threshold.

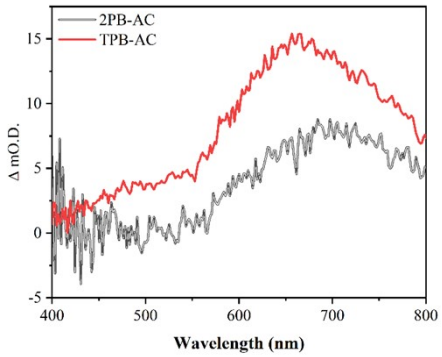

Fig. S9 Triplet absorption spectra of 2PB-AC and TPB-AC, measured at the degassed solutions (excitation wavelength: 343 nm)

Table S1. Energy levels of singlet and triplet states and oscillator strengths of singlet states in 2PB-AC and TPB-AC

Excited States	2PB-AC			TPB-AC		
	S energy level (eV)	T energy level (eV)	S Oscillator Strength	S energy level (eV)	T energy level (eV)	S Oscillator Strength
1	2.9943	2.5691	0.5113	3.0491	2.6976	0.2847
2	3.6246	3.0705	0.0123	3.5404	3.0698	0.0060
3	3.7940	3.1453	0.3832	3.6445	3.1354	0.0132
4	3.8925	3.3044	0.1791	3.7316	3.2811	0.3634
5	3.9917	3.3533	0.0009	3.8920	3.2988	0.1238
6	4.1090	3.6027	0.0011	3.9147	3.3387	0.1606
7	4.1639	3.7984	0.5949	4.0006	3.5468	0.0090
8	4.2986	3.8533	0.0239	4.1890	3.6422	0.0285
9	4.3099	3.8808	0.0045	4.2946	3.6930	0.0114
10	4.6017	3.9009	0.0010	4.3071	3.8146	0.1362

Table S2. Photophysical parameters of 2PB-AC and TPB-AC in different solvents.

	2PB-AC						TPB-AC					
	Abs /nm	λ_{PL}[(a)] /nm	fwhm[(b)] /nm	τ[(c)] /ns	Φ_{PL}[(d)]	k_r[(e)] /ns^{-1}	Abs /nm	λ_{PL}[(a)] /nm	fwhm[(b)] /nm	τ[(c)] /ns	Φ_{PL}[(d)]	k_r[(e)] /ns^{-1}
HEX	354	408	44	1.37	0.71	0.52	344	408	56	1.56	0.66	0.42
TOL	359	436	59	1.60	0.85	0.53	347	437	63	2.12	0.81	0.38
THF	355	474	88	2.43	0.85	0.35	343	483	90	3.44	0.85	0.25
DCM	356	491	96	2.82	0.97	0.34	343	495	97	4.08	0.94	0.23
DMF	351	527	132	3.54	0.82	0.23	339	544	135	5.28	0.77	0.15

[(a)] Peak wavelength of the PL spectrum, excitation wavelength at 355 nm; [(b)] full width at half-maximum of the PL spectrum; [(c)] Excitation wavelength at 375 nm and probe wavelength at the peak wavelength of the PL spectrum; [(d)] Excitation wavelength at 330 nm; [(e)] $k_r=\Phi_{PL}/\tau$.

Table S3. Photophysical parameters of 2PB-AC and TPB-AC films in different doping concentration.

2PB-AC						TPB-AC					
C[(a)] /%	λ_{PL}[(b)] /nm	Φ_{PL}[(c)] %	τ[(d)] (ns)	kr/ ns^{-1}	knr*10 /ns^{-1}	C[(a)] /%	λ_{PL}[(b)] /nm	Φ_{PL}[(c)] %	τ[(d)] (ns)	kr/ ns^{-1}	knr*10/n s^{-1}
2	445	97	1.89	0.51	0.16	1	425	99	2.10	0.47	0.38
4	449	97	1.97	0.49	0.15	3	429	99	2.17	0.46	0.09
6	451	97	2.05	0.47	0.15	5	432	99	2.24	0.45	0.04
8	453	96	2.06	0.47	0.19	7	433	99	2.28	0.44	0.22
12	457	95	2.10	0.45	0.24	9	439	98	2.35	0.42	0.47
16	464	95	2.29	0.42	0.22	11	442	97	2.42	0.40	1.24

20	466	97	2.82[e] 2.15(0.74) 4.71(0.26)	0.34	0.11	17	444	99	2.45	0.40	0.57
30	470	95	3.02[e] 1.99(0.52) 4.13(0.48)	0.31	0.17	23	445	98	2.46	0.40	0.89
40	478	96	3.62[e] 2.55(0.71) 6.29(0.29)	0.27	0.11	29	445	97	2.54	0.38	1.26
50	483	95	3.79[e] 2.5(0.63) 5.96(0.37)	0.25	0.13	50	449	99	2.73	0.36	0.48
70	488	94	4.07[e] 2.36(0.45) 5.55(0.54)	0.23	0.15	70	452	98	3.15[e] 2.43(0.72) 4.98(0.28)	0.31	0.41
100	493	93	5.51[e] 3.30(0.72) 10.2(0.28)	0.17	0.15	100	455	98	3.43[e] 2.5(0.69) 5.49(0.31)	0.29	0.58

[a] Doping concentration; [b], [c] Excitation wavelength at 330 nm; [d] Excitation wavelength at 375 nm and probe wavelength at the peak wavelength of the PL spectrum; [e] Average lifetime, $\tau_{AVE} = \sum A_i\tau_i^2 / \sum A_i\tau_i$.

Table S4. ASE performance parameters of 2PB-AC: CBP (x: 100-x, in weight) doped films.

Doping ratio (%)	E_{th} (μJ cm^{-2})	λ_{ASE} (nm)	FWHM (nm)
1	3.85	444	7.7
3	2.45	450	7.7
5	1.31	453	6.6
7	1.06	453	7.0
9	1.21	455	5.9
17	3.91	460	7.3
23	4.41	461	6.6
29	5.56	462	6.9
33	8.49	462	6.6
50	8.45	463	5.5
70	27.14	464	5.1

Table S5. ASE performance parameters of 2PB-AC in different solvents.

	a[1]	**b**	**c**	**d**	**e**	**f**
λ_{ASE}/(nm)	408	416	430	439	446	456
FWHM/(nm)	1.8	1.8	3.7	4.4	3.6	8.8
E_{th} (mJ cm^{-2})	29.2	1.14	1.62	1.95	3.83	>100

[1] Saturated solution.

Reference

[1] T. Lu, F. W. Chen. Multiwfn: A multifunctional wavefunction analyzer. *J. Comput. Chem.*, **2012**, 33(5):580-92.