Table S1 Selected bond lengths (Angstroms), bond angles (degrees), and dihedral angles (degrees) (computed using the B3LYP functional) for the Cu(I) dyes, in solvent. Expt denotes the experimental values (Ref. 11,14) for the corresponding homoleptic dyes of type $[CuL_2]^+$, where L=6,6'-dimethyl-2,2'-bipyridine-dimethylformate).

Parameter	Dye1	Dye2	Dye3	Dye4	Dye5	Dye6	expt (Ref. 11,14)
Cu-N1	2.08	2.08	2.09	2.09	2.08	2.09	2.01
Cu-N2	2.08	2.08	2.09	2.08	2.08	2.08	2.01
Cu-N3	2.08	2.07	2.09	2.09	2.10	2.09	2.04
Cu-N4	2.08	2.07	2.09	2.09	2.10	2.10	2.00
N1-Cu-N2	80.08	80.24	80.34	80.37	80.33	80.37	80.93
N2-Cu-N3	125.99	125.55	119.92	119.84	119.72	120.18	119.26
N3-Cu-N4	81.11	80.78	81.09	81.14	80.95	80.01	81.21
N1-Cu-N4	125.95	126.03	119.39	119.09	119.64	118.71	134.76
N2-Cu-N4	125.25	125.64	131.88	131.87	131.90	132.01	122.14
N1-Cu-N3	125.05	125.26	131.70	132.04	131.80	132.13	123.77
τ_4	0.77	0.79	0.68	0.68	0.68	0.68	-

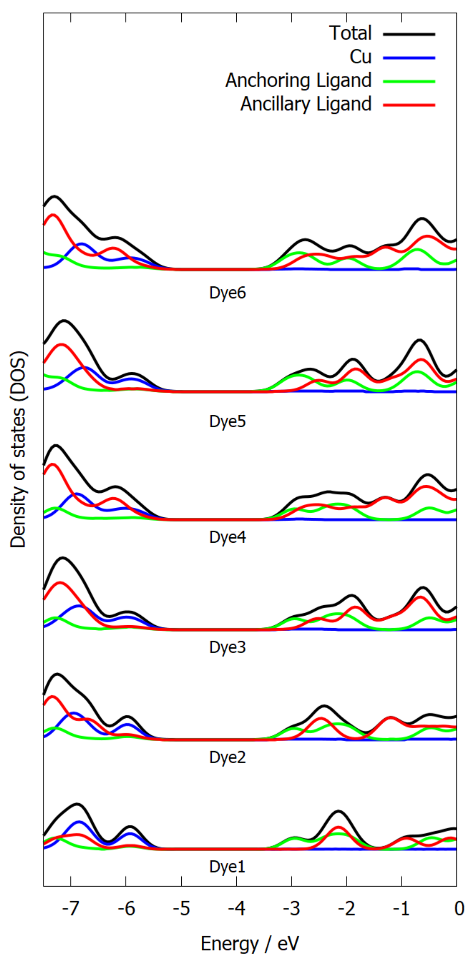

Figure S1 Projected Density of States (PDOS)(using the B3LYP functional) of the Cu(I) dyes, in solvent.

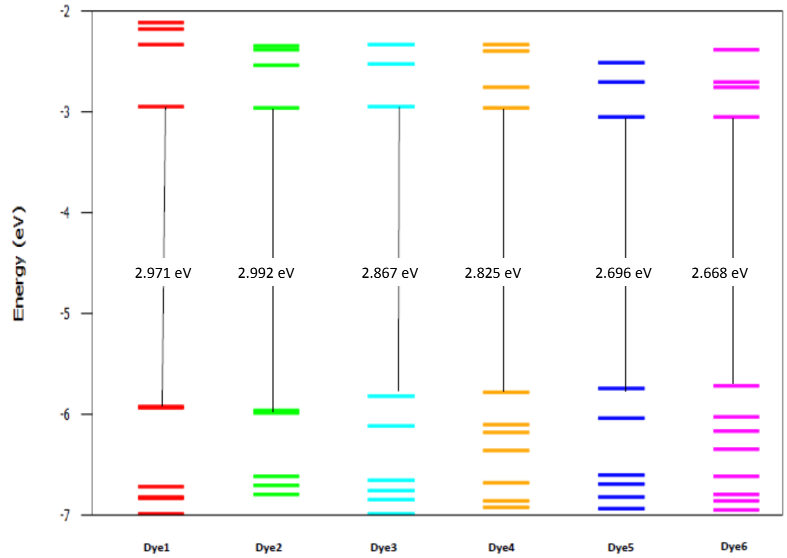

Figure S2 Kohn- Sham energy levels (using the B3LYP functional) for the Cu(I) dyes, in solvent.

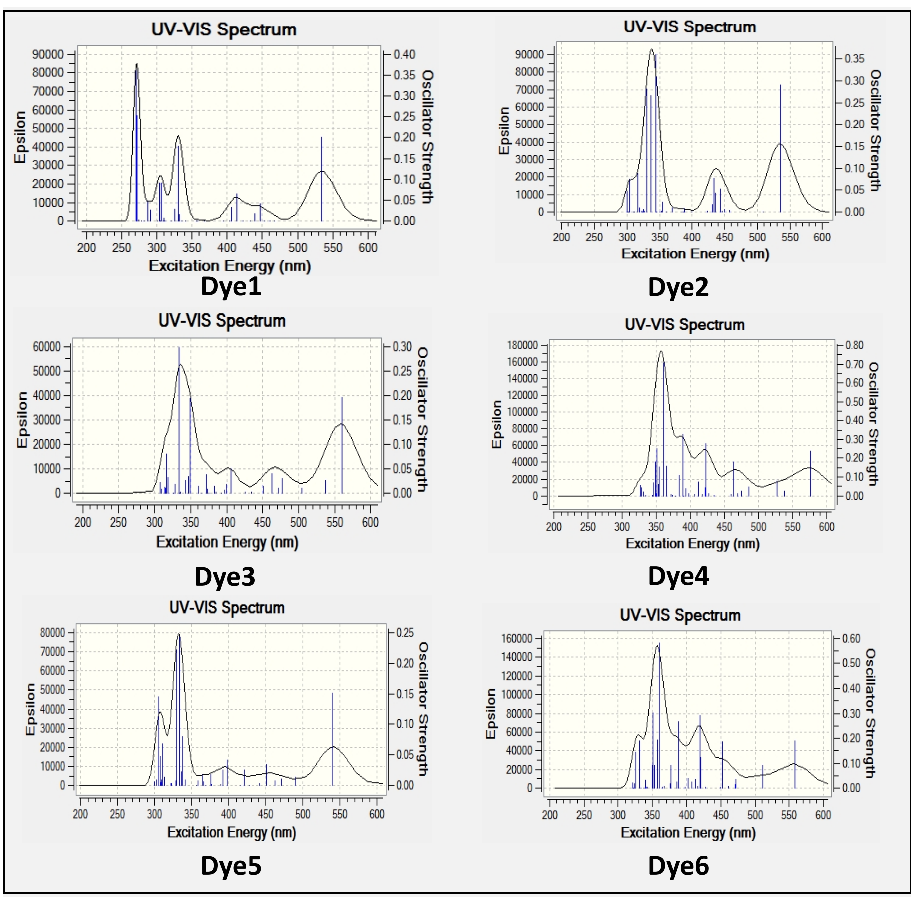

Figure S3 Optical Absorption Spectra (using the B3LYP functional) of the Cu(I) dyes, in solvent.

Table S2 Optical data corresponding to the theoretical absorption spectra (using the B3LYP functional) for the Cu(I) dyes, in solvent. ESN denotes the transition from the single ground state S_0 to the singlet excited state S_N. Only the transitions with oscillator strength f $\geq$ 0.1 have been included in the table.

System	Transitions	Excitation energy(eV)	Wavelength(nm)(λ)	Oscillator strength (f)
Dye1	ES2($S_0 \rightarrow S_2$)	2.32	533.95	0.20
	ES10($S_0 \rightarrow S_{10}$)	2.99	413.88	0.06
	ES21($S_0 \rightarrow S_{21}$)	3.74	331.02	0.18
	ES22($S_0 \rightarrow S_{22}$)	3.74	330.77	0.11
Dye2	ES2($S_0 \rightarrow S_2$)	2.31	534.88	0.29
	ES9($S_0 \rightarrow S_9$)	2.85	433.65	0.08
	ES22($S_0 \rightarrow S_{22}$)	3.59	344.64	0.36
	ES23($S_0 \rightarrow S_{23}$)	3.68	336.75	0.27
	ES24($S_0 \rightarrow S_{24}$)	3.75	330.19	0.28
Dye3	ES1($S_0 \rightarrow S_1$)	1.84	672.74	0.04
	ES2($S_0 \rightarrow S_2$)	2.21	559.77	0.20
	ES23($S_0 \rightarrow S_{23}$)	3.55	349.25	0.19
	ES28($S_0 \rightarrow S_{28}$)	3.71	333.67	0.30
Dye4	ES1($S_0 \rightarrow S_1$)	1.84	673.53	0.07
	ES2($S_0 \rightarrow S_2$)	2.15	575.64	0.24
	ES4($S_0 \rightarrow S_4$)	2.35	526.83	0.08
	ES8($S_0 \rightarrow S_8$)	2.67	463.25	0.18
	ES11($S_0 \rightarrow S_{11}$)	2.90	426.15	0.01
	ES12($S_0 \rightarrow S_{12}$)	2.93	422.96	0.28
	ES21($S_0 \rightarrow S_{21}$)	3.19	388.39	0.32
	ES23($S_0 \rightarrow S_{23}$)	3.23	383.53	0.11
	ES27($S_0 \rightarrow S_{27}$)	3.39	365.40	0.16
	ES28($S_0 \rightarrow S_{28}$)	3.44	360.39	0.71
	ES29($S_0 \rightarrow S_{29}$)	3.50	353.54	0.15
	ES31($S_0 \rightarrow S_{31}$)	3.53	350.27	0.25
	ES33($S_0 \rightarrow S_{33}$)	3.56	348.12	0.18
Dye5	ES1($S_0 \rightarrow S_1$)	1.90	649.22	0.02
	ES2($S_0 \rightarrow S_2$)	2.29	540.39	0.15
	ES22($S_0 \rightarrow S_{22}$)	3.67	337.33	0.19
	ES26($S_0 \rightarrow S_{26}$)	3.76	329.65	0.04
Dye6	ES1($S_0 \rightarrow S_1$)	1.90	650.20	0.06
	ES2($S_0 \rightarrow S_2$)	2.22	558.49	0.19
	ES7($S_0 \rightarrow S_7$)	2.74	452.43	0.19
	ES10($S_0 \rightarrow S_{10}$)	2.94	420.91	0.12
	ES11($S_0 \rightarrow S_{11}$)	2.95	419.50	0.29
	ES18($S_0 \rightarrow S_{18}$)	3.19	387.65	0.27
	ES20($S_0 \rightarrow S_{20}$)	3.23	383.53	0.11
	ES24($S_0 \rightarrow S_{24}$)	3.39	365.40	0.16
	ES25($S_0 \rightarrow S_{25}$)	3.44	360.39	0.71
	ES26($S_0 \rightarrow S_{26}$)	3.50	353.54	0.15
	ES27($S_0 \rightarrow S_{27}$)	3.51	352.53	0.06

Table S3 Optical data corresponding to the theoretical absorption spectra (using the CAM-B3LYP functional) of the Cu(I) dyes, in solvent. ESN denotes the transition from the singlet ground state S_0 to the singlet excited state S_N. Only the transitions with oscillator strength f$\geq$ 0.1 have been included in the table.

System	Transitions	Excitation energy(eV)	Wavelength(nm)(λ)	Oscillator strength (f)
Dye1	ES1($S_0 \rightarrow S_1$)	2.67	462.80	0.00
	ES2($S_0 \rightarrow S_2$)	3.08	402.94	0.20
	ES13($S_0 \rightarrow S_{13}$)	4.20	295.15	0.40
	ES24($S_0 \rightarrow S_{24}$)	4.92	251.75	0.75
Dye2	ES1($S_0 \rightarrow S_1$)	2.72	455.20	0.00
	ES2($S_0 \rightarrow S_2$)	3.09	400.81	0.29
	ES9($S_0 \rightarrow S_9$)	3.86	320.92	0.19
	ES16($S_0 \rightarrow S_{16}$)	4.20	295.16	0.40
	ES19($S_0 \rightarrow S_{22}$)	4.34	285.67	0.31
	ES20($S_0 \rightarrow S_{22}$)	4.47	277.08	0.43
	ES22($S_0 \rightarrow S_{22}$)	4.68	264.98	0.28
Dye2a	ES1($S_0 \rightarrow S_1$)	2.31	535.51	0.00
	ES2($S_0 \rightarrow S_2$)	3.06	404.74	0.29
	ES5($S_0 \rightarrow S_5$)	3.62	342.37	0.14
	ES8($S_0 \rightarrow S_8$)	3.78	327.77	0.12
	ES9($S_0 \rightarrow S_9$)	3.85	322.15	0.18
	ES15($S_0 \rightarrow S_{15}$)	4.20	295.02	0.38
	ES19($S_0 \rightarrow S_{19}$)	4.33	285.90	0.25
	ES24($S_0 \rightarrow S_{24}$)	4.76	260.17	0.53
Dye2b	ES1($S_0 \rightarrow S_1$)	2.44	508.03	0.00
	ES3($S_0 \rightarrow S_3$)	3.23	383.83	0.13
	ES8($S_0 \rightarrow S_8$)	3.78	327.83	0.11
	ES15($S_0 \rightarrow S_{15}$)	4.19	296.07	0.17
	ES17($S_0 \rightarrow S_{17}$)	4.21	294.60	0.30
	ES18($S_0 \rightarrow S_{18}$)	4.22	293.99	0.28
	ES19($S_0 \rightarrow S_{19}$)	4.37	283.35	0.56
	ES22($S_0 \rightarrow S_{22}$)	4.49	275.98	0.40
	ES31($S_0 \rightarrow S_{31}$)	4.92	251.66	0.30
Dye3	ES1($S_0 \rightarrow S_1$)	2.66	464.64	0.03
	ES2($S_0 \rightarrow S_2$)	3.07	403.98	0.13
	ES11($S_0 \rightarrow S_{11}$)	4.09	303.41	0.18
	ES15($S_0 \rightarrow S_{15}$)	4.20	295.43	0.32
	ES17($S_0 \rightarrow S_{17}$)	4.27	290.19	0.36
	ES25($S_0 \rightarrow S_{25}$)	4.80	258.19	0.60
Dye4	ES1($S_0 \rightarrow S_1$)	2.67	462.67	0.06
	ES2($S_0 \rightarrow S_2$)	3.01	411.94	0.19
	ES3($S_0 \rightarrow S_3$)	3.24	382.83	0.15
	ES4($S_0 \rightarrow S_4$)	3.47	356.98	0.21
	ES5($S_0 \rightarrow S_5$)	3.55	348.88	0.60
	ES6($S_0 \rightarrow S_6$)	3.57	347.00	0.46
	ES10($S_0 \rightarrow S_{10}$)	3.83	323.36	0.97
	ES16($S_0 \rightarrow S_{16}$)	4.13	300.47	0.20
	ES18($S_0 \rightarrow S_{18}$)	4.19	295.69	0.13
	ES19($S_0 \rightarrow S_{19}$)	4.21	294.78	0.25
	ES24($S_0 \rightarrow S_{24}$)	4.71	263.03	0.17
	ES34($S_0 \rightarrow S_{34}$)	4.96	249.75	0.30
Dye5	ES1($S_0 \rightarrow S_1$)	2.70	459.09	0.05
	ES2($S_0 \rightarrow S_2$)	3.04	407.22	0.14
	ES12($S_0 \rightarrow S_{12}$)	4.02	308.03	0.13
	ES15($S_0 \rightarrow S_{15}$)	4.14	299.41	0.28
	ES17($S_0 \rightarrow S_{17}$)	4.26	291.06	0.40
	ES23($S_0 \rightarrow S_{23}$)	4.71	263.11	0.72
	ES25($S_0 \rightarrow S_{25}$)	4.78	259.30	0.39
Dye6	ES1($S_0 \rightarrow S_1$)	2.70	458.24	0.11
	ES2($S_0 \rightarrow S_2$)	2.98	416.26	0.16
	ES3($S_0 \rightarrow S_3$)	3.21	385.65	0.27
	ES5($S_0 \rightarrow S_5$)	3.55	348.85	0.99
	ES9($S_0 \rightarrow S_9$)	3.82	324.09	1.07
	ES17($S_0 \rightarrow S_{17}$)	4.09	303.39	0.13
	ES18($S_0 \rightarrow S_{18}$)	4.14	299.19	0.29
	ES25($S_0 \rightarrow S_{25}$)	4.63	267.69	0.27
	ES40($S_0 \rightarrow S_{40}$)	4.99	249.53	0.29

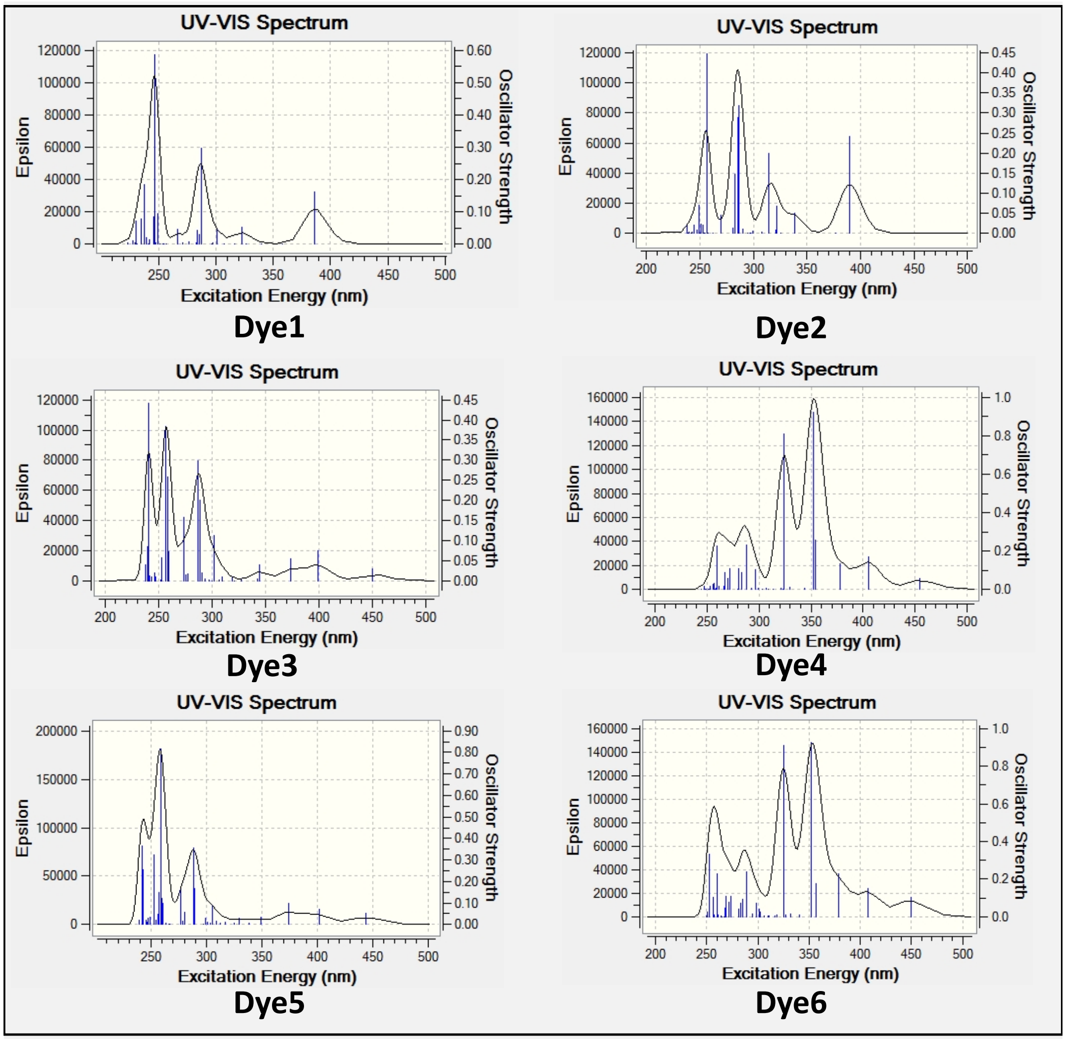

Figure S4 Optical Absorption Spectra (using the CAM-B3LYP functional) for the Cu(I) dyes, in gas phase.

Table S4 Optical data corresponding to the theoretical absorption spectra (using the CAM-B3LYP functional) for the Cu(I) dyes, in gas phase. ESN denotes the transition from the single ground state S_0 to the singlet excited state S_N. Only the transitions with oscillator strength f $\geq$ 0.1 have been included in the table.

System	Transitions	Excitation energy(eV)	Wavelength(nm)(λ)	Oscillator strength (f)
Dye1	ES1($S_0 \rightarrow S_1$)	2.79	443.33	0.00
	ES2($S_0 \rightarrow S_2$)	3.21	386.57	0.16
	ES13($S_0 \rightarrow S_{13}$)	4.32	287.05	0.30
	ES27($S_0 \rightarrow S_{27}$)	5.03	246.57	0.58
	ES33($S_0 \rightarrow S_{33}$)	5.22	237.50	0.18
Dye2	ES1($S_0 \rightarrow S_1$)	2.79	443.29	0.00
	ES2($S_0 \rightarrow S_2$)	3.18	390.05	0.24
	ES9($S_0 \rightarrow S_9$)	3.94	314.41	0.20
	ES16($S_0 \rightarrow S_{16}$)	4.32	286.93	0.32
	ES17($S_0 \rightarrow S_{17}$)	4.34	285.69	0.28
	ES18($S_0 \rightarrow S_{18}$)	4.38	283.10	0.14
	ES24($S_0 \rightarrow S_{24}$)	4.84	256.37	0.44
Dye3	ES1($S_0 \rightarrow S_1$)	2.75	450.23	0.03
	ES11($S_0 \rightarrow S_{11}$)	4.10	302.23	0.11
	ES16($S_0 \rightarrow S_{16}$)	4.29	288.80	0.20
	ES17($S_0 \rightarrow S_{17}$)	4.32	286.94	0.30
	ES20($S_0 \rightarrow S_{20}$)	4.53	273.63	0.16
	ES22($S_0 \rightarrow S_{22}$)	4.79	258.81	0.25
	ES25($S_0 \rightarrow S_{25}$)	4.83	256.48	0.38
	ES36($S_0 \rightarrow S_{36}$)	5.13	241.26	0.44
Dye4	ES1($S_0 \rightarrow S_1$)	2.72	454.85	0.06
	ES2($S_0 \rightarrow S_2$)	3.06	405.54	0.17
	ES3($S_0 \rightarrow S_3$)	3.28	377.85	0.14
	ES4($S_0 \rightarrow S_4$)	3.50	354.24	0.26
	ES5($S_0 \rightarrow S_5$)	3.52	352.64	0.92
	ES8($S_0 \rightarrow S_8$)	3.82	324.18	0.81
	ES16($S_0 \rightarrow S_{16}$)	4.18	296.60	0.10
	ES19($S_0 \rightarrow S_{19}$)	4.31	287.93	0.23
	ES21($S_0 \rightarrow S_{21}$)	4.42	280.20	0.11
	ES23($S_0 \rightarrow S_{23}$)	4.56	271.72	0.11
	ES28($S_0 \rightarrow S_{28}$)	4.76	260.16	0.22
Dye5	ES1($S_0 \rightarrow S_1$)	2.78	444.42	0.05
	ES16($S_0 \rightarrow S_{16}$)	4.28	289.34	0.17
	ES17($S_0 \rightarrow S_{17}$)	4.30	288.31	0.36
	ES20($S_0 \rightarrow S_{20}$)	4.48	276.75	0.16
	ES25($S_0 \rightarrow S_{25}$)	4.76	259.94	0.12
	ES26($S_0 \rightarrow S_{26}$)	4.78	259.17	0.81
	ES27($S_0 \rightarrow S_{27}$)	4.82	256.85	0.14
	ES30($S_0 \rightarrow S_{30}$)	4.91	252.28	0.32
	ES37($S_0 \rightarrow S_{37}$)	5.10	242.67	0.26
	ES38($S_0 \rightarrow S_{38}$)	5.12	242.13	0.36
Dye6	ES1($S_0 \rightarrow S_1$)	2.75	449.25	0.10
	ES1($S_0 \rightarrow S_1$)	2.76	449.25	0.10
	ES2($S_0 \rightarrow S_2$)	3.04	407.63	0.15
	ES3($S_0 \rightarrow S_3$)	3.27	378.74	0.23
	ES4($S_0 \rightarrow S_4$)	3.47	357.02	0.18
	ES5($S_0 \rightarrow S_5$)	3.52	352.51	0.93
	ES9($S_0 \rightarrow S_9$)	3.82	324.91	0.91
	ES19($S_0 \rightarrow S_{19}$)	4.29	288.90	0.24
	ES23($S_0 \rightarrow S_{23}$)	4.53	273.14	0.10
	ES25($S_0 \rightarrow S_{25}$)	4.61	268.48	0.10
	ES31($S_0 \rightarrow S_{31}$)	4.75	260.48	0.16
	ES32($S_0 \rightarrow S_{32}$)	4.77	259.88	0.23
	ES35($S_0 \rightarrow S_{35}$)	4.83	256.66	0.10
	ES37($S_0 \rightarrow S_{37}$)	4.91	252.49	0.33

Table S5 Computed light harvesting efficiency (LHE) (using the B3LYP functional), in the first two singlet excited states, for the Cu(I) dyes, in solvent.

Copper (I) Dye	f_1	LHE_1	f_2	LHE_2	Average LHE
Dye1	0.00	0.00	0.20	0.37	0.18
Dye2	0.00	0.00	0.29	0.49	0.24
Dye3	0.04	0.08	0.20	0.36	0.22
Dye4	0.07	0.15	0.24	0.42	0.29
Dye5	0.02	0.06	0.15	0.29	0.18
Dye6	0.06	0.12	0.19	0.35	0.24

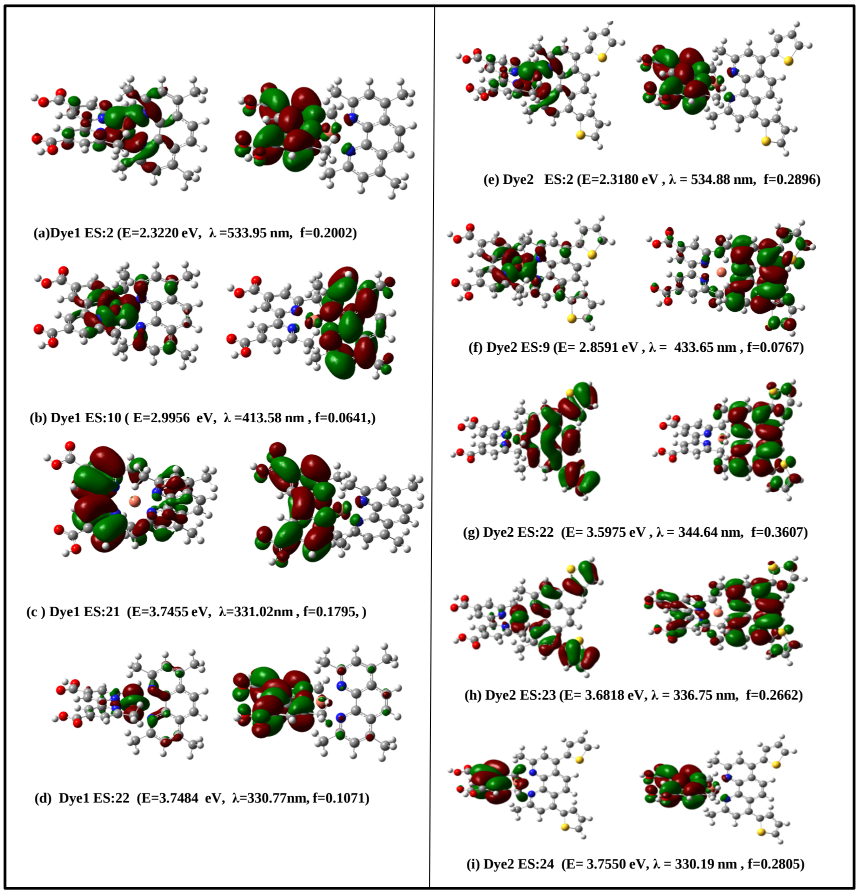

Figure S5 Natural transition orbitals (NTOs) (using the B3LYP functional), showing the charge transfer upon light absorption, for the Cu(I) dyes, Dye1 (left panel) and Dye2 (right panel), in solvent.

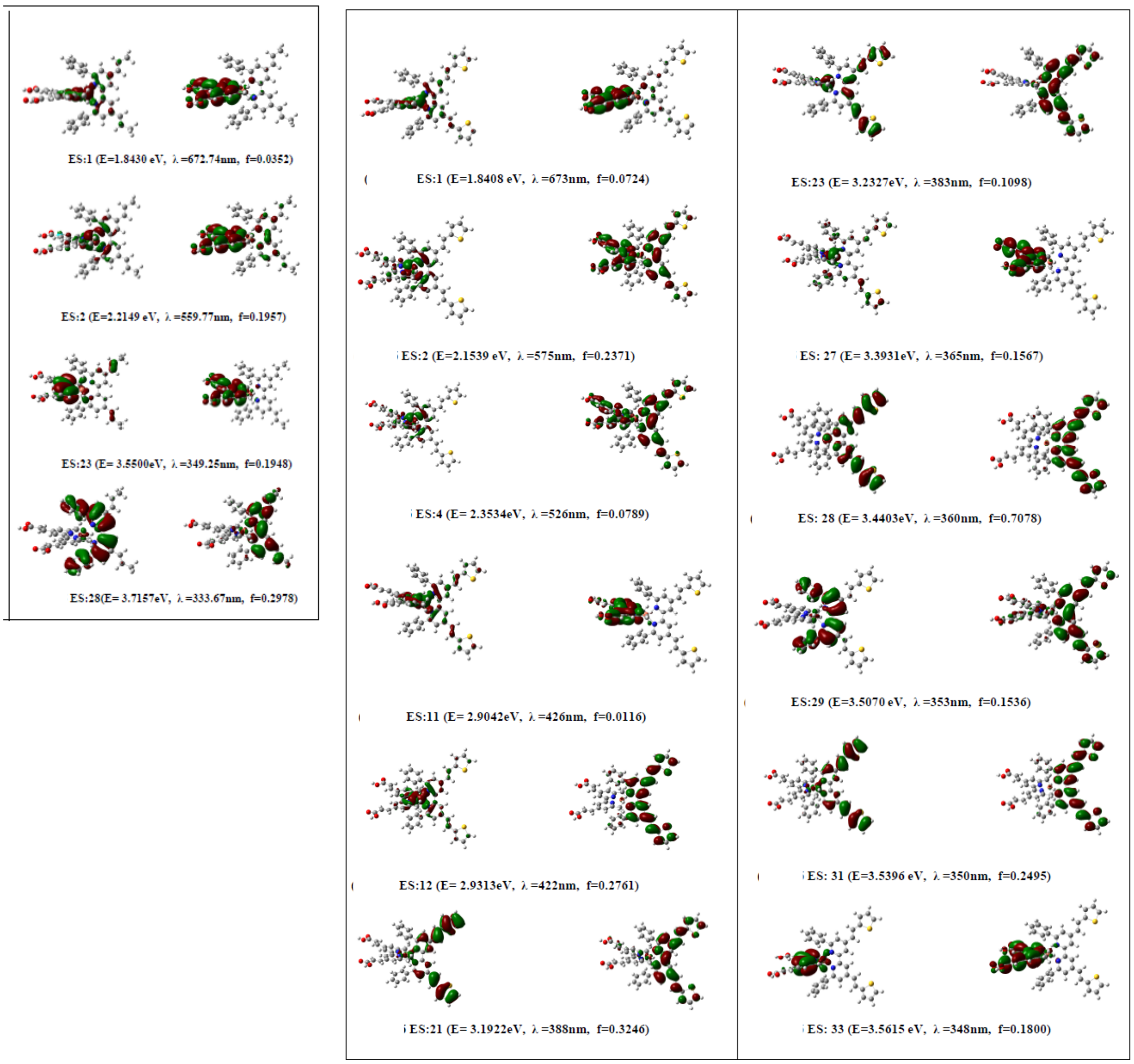

Figure S6 Natural transition orbitals (NTOs) (using the B3LYP functional), showing the charge transfer upon light absorption, for the Cu(I) dyes, Dye3 (left panel) and Dye4 (right panel), in solvent.

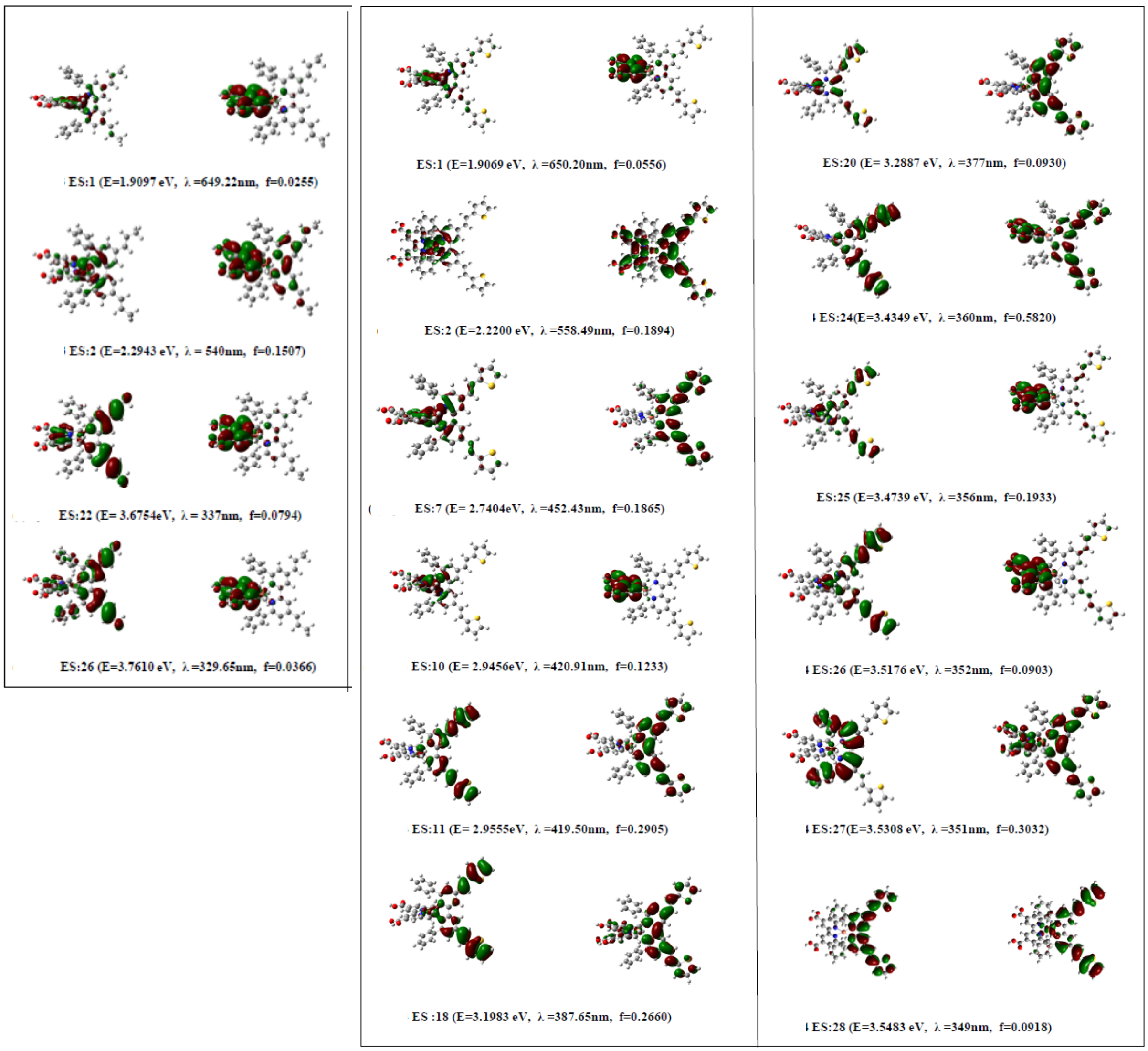

Figure S7 Natural transition orbitals (NTOs) (using the B3LYP functional), showing the charge transfer upon light absorption, for the Cu(I) dyes, Dye5 (left panel) and Dye6 (right panel), in solvent.

Table S6 Percentage contributions of the orbitals involved in the charge transfer upon light absorption (using the CAM-B3LYP functional) for the Cu(I) dyes, in solvent.

Dyes	excited state	Hole											Particle												
		Metal			Anchoring ligand				Ancillary ligand				Metal			Anchoring ligand					Ancillary ligand				
		Cu-s	Cu-p	Cu-d	C-s	C-p	N-s	N-p	C-s	C-p	N-s	N-p	Cu-s	Cu-p	Cu-d	C-s	C-p	N-s	N-p	O-p	C-s	C-p	N-s	N-p	S-p
Dye1	ES1		2.19	73.62		2.19	4.39	17.58							3.44		57.47		32.18	4.59		2.29			
	ES2			80.8		2.1				2.1		14.9			2.2		66.7		28	3.2					
	ES13			2.2		97.8									2.2		66.3		26.1	5.4					
	ES19			15.2		76.1		8.7														100			
	ES24			9.67						90.32					1.09							74.72		24.17	
Dye2	ES1		2.15	72.04		2.15	4.30	17.20							3.22		64.51		30.11			2.15			
	ES2			82.6		2.2					4.3	10.9			4.1		63.9		26.8	3.1		2.1			
	ES9			67.8				16.1		14.9		1.1					10		4.3			73.9		11.6	
	ES16			3.2		96.8									2.2		66.7		25.8	5.4					
	ES19		1.1	24.2						65.9		8.8										88.6		9.1	2.3
	ES20			8.3						85.4		6.2										85.4		10.1	4.5
	ES22			2.15						97.84					1.11							75.55		22.22	1.11
Dye2a	ES1			77.01			4.59	18.39									59.34		30.76	3.29		6.59			
	ES2		3.3	79.3		2.2					4.3	10.9			2.1		65.3		27.4	3.2		2.1			
	ES5			63.2				11.5		21.8		3.4			1.1							75.3		23.6	
	ES8		3.5	61.2						12.9		22.4										82.7		17.3	
	ES9		1.1	61.4			2.3	6.8		20.4		8					11.5		5.1			79.5		3.8	
	ES15			2.2		97.8									2.2		60.2		25.8	5.4		6.4			
	ES19			48.91						51.08					2.40		67.46		28.91	6.02		7.22			
	ES24			17.5		82.5									2.15							73.11		24.73	
Dye2b	ES1		2.32	76.74			4.65	16.27									60.67		29.21	1.12		8.98			
	ES3		2.1	78.7		2.1		14.9		2.1					3.2		45.7		21.3	1.1		20.2		8.5	
	ES8			67.1				7.1		18.8	1.1	7.1										88.6	16.4		
	ES15			36			1.2	7		48.8		7					2.6					85.9		11.5	
	ES17			30.2				5.8		59.3		7										90.4		9.6	
	ES18		4.2	70.8		10.4		4.2		2.1		8.3					62.4		18.3	8.6		10.8			
	ES19			5.37						94.62												95.40		4.59	
	ES22			50		45.65		4.34									98.85					1.14			
	ES31			18.88		73.33		2.22				6.66										95.65		4.34	
Dye3	ES1		2.98	77.01			2.29	18.39							2.19		68.13		28.57	1.09					
	ES2		3.2	75.3						4.3	4.3	12.9			1		28.6		6.1		2	49.9		13.3	
	ES11		5	74.1				4.7		1.2	2.4	14.1										82.2		11.8	
	ES15		3.2	96.8											2.1		61.7		23.4			12.8			
	ES17			44.9						55.1				4.1	2							69.4		24.5	
	ES25					70.73	6.09			20.73		2.43			3		37		11			42		7	
Dye4	ES1		2.32	76.74			2.32	16.27				2.32			2.32		61.62		27.90	2.32		5.81			
	ES2		3.4	74.2		1.1	2.2	7.9							1.2		34.1		16.5			36.5		11.8	
	ES3		2.2	80.2		2.2	2.2	5.5			2.2	5.5			3.6		52.4		24.4			12.2		7.3	
	ES4			79.3		1.2	2.4	11		4.9		1.2					23.6		13.9			48.6		13.9	
	ES5	2.1	5.3	88.3										1	3.2	1	60		27.4	5.3	1	1			
	ES6	2.3	6.8	85.2				3.4		1.1		1.1			2.2		62.2		25.6	2.2	1.1	5.6		1.1	
	ES10			18		5.6		1.1		75.3					1.1		18.9		15.6			60			4.4
	ES16			41.3		26.1				32.6				2.2			26.7		12.2			46.7		12.2	
	ES18			84.9		4.3				10.8			2.3			29.1		14			40.7		14		
	ES19			3.3		96.7								2.1			68.1		23.4	5.3		1.1			
	ES24					90.58		9.41									20.51					79.48			
	ES34									45		55										87.35		3.44	9.19
Dye5	ES1		2.27	75				20.45				2.87			2.70		55.90		31.08			10.81			
	ES2		3.2						4.3	4.3	15.1			1.1		8.4		6.3			63.2		21.1		
	ES12		3.5	75.3				4.7		2.4	2.4	11.8									1.1	81.5		17.4	
	ES15			2.1		97.8									2		72.7		20.2	5					
	ES17			10.8		89.2									1							76.3		22.7	
	ES20			80				88.9		33.2		77.8									3.3	79.8		16.8	
	ES23			43.33		40		2.22		14.44							42.85		10.98	4.39		41.75			
	ES25				3.90	26.13		9.09		64.77							43.82		16.85	4.49		34.83			
Dye6	ES1		2.4	78.6			2.4	14.3				2.4			2.3		53.4		25			13.6		2.3	
	ES2		3.4	73.9						2.3	4.5	15.9			2.3		2.3					70.4		25	
	ES3			52.2		17.4		2.2		22.8		5.4		1	80.2				1			2.2	2.2	13.2	
	ES5			10.5		41.9				40.7		7			1		15.4					63.7		15.4	4.4
	ES9			12.9		44.7				38.8		3.6			1.2		18.8					60		15.3	4.7
	ES17	1.1		75		23.9									2.3		2.3					67.8		27.6	
	ES18			2.2		97.8									2		56.2		21.8	2		17.7			
	ES25			33.69		58.69		5.43		2.17							48.38		10.75	4.30		36.55			
	ES40									29.48		70.57										90.69		8.13	1.16