Supporting information

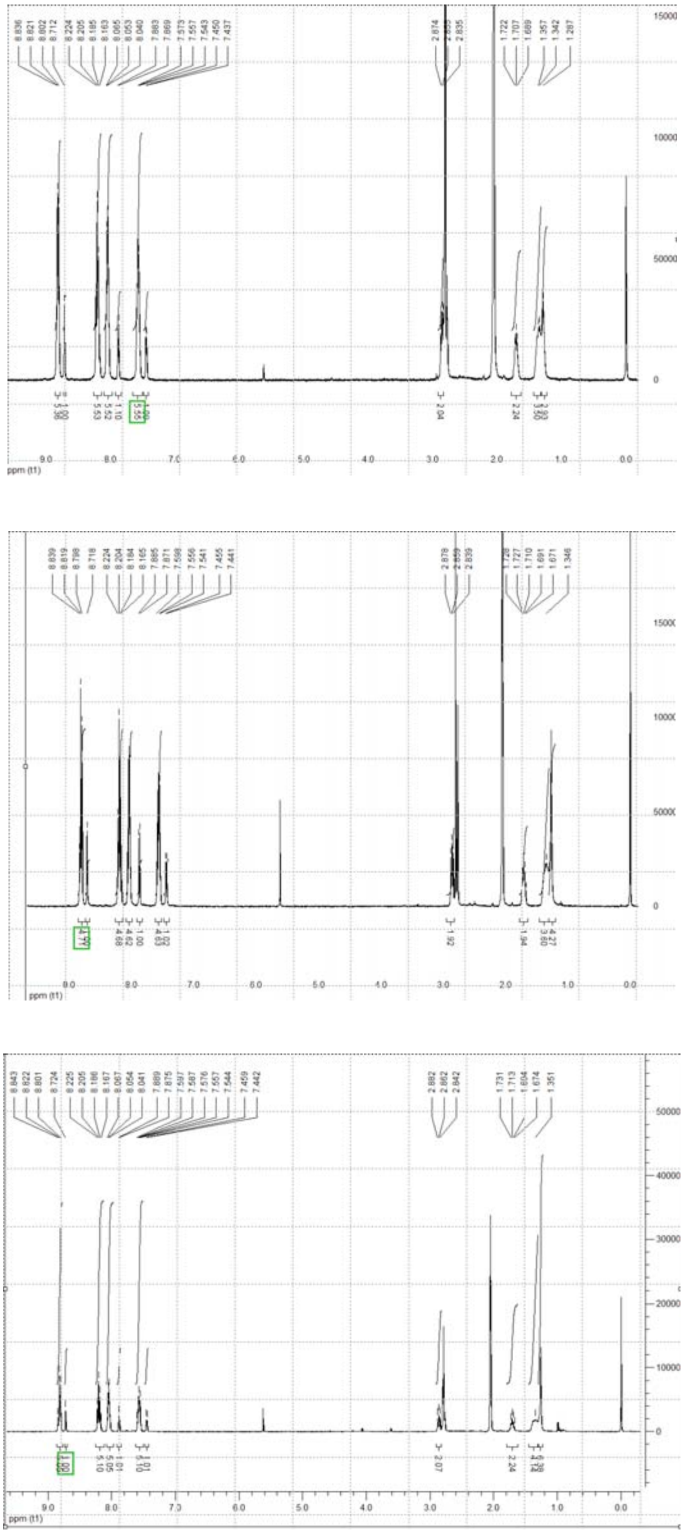

Fig. S1 ^{1}H NMR spectra for bimetallic ruthenium complexes **1**, from top to down, **1a**, **1b** and **1c** respectively.

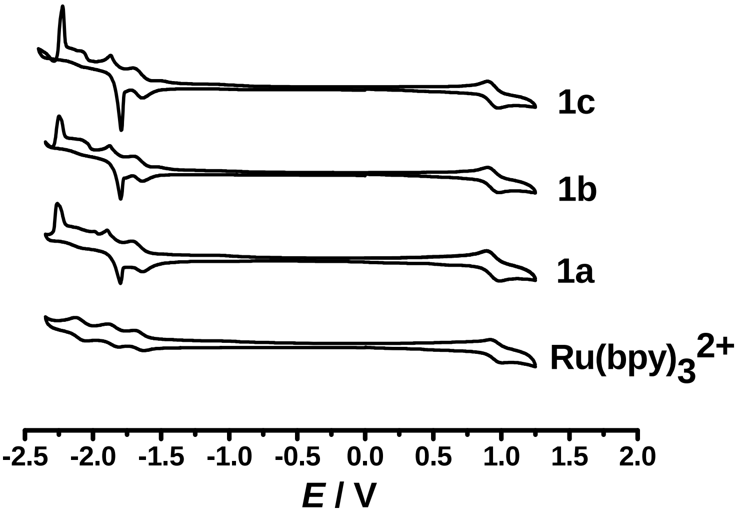

Fig. S2 Cyclic voltammetry (CV) of 1 μM bimetallic ruthenium complexes **1** in PF_6^- salt at glassy carbon electrode, scan rate: 0.1 V/s

Table S1. Selected Electronic Excitation Energies (eV) and Oscillator Strengths (*f*), Configurations of the Low-Lying Excited States of the complexes **1**[a]

dyes	Electronic transition	Energy[b] (ev)	*f*[c]	Composition[d]	CI[e]
1d	S_0-S_{11}	2.7549 (450.4 nm)	0.0110	H-2→L+1	0.55
				H-2→L+5	0.43
	S_0-S_{12}	2.7559 (450.0 nm)	0.0164	H-3→L	0.52
				H-3→L+4	0.46
	S_0-S_{13}	2.8623 (433.2 nm)	0.1187	H-5→L+2	0.40
				H-3→L	0.33
				H-3→L+4	0.39
	S_0-S_{14}	2.8726 (431.6 nm)	0.1309	H-4→L+3	0.43
				H-2→L+1	0.34
				H-2→L+5	0.39
	S_0-S_{15}	2.8865 (429.5 nm)	0.1338	H-5→L	0.29
				H-5→L+4	0.38
				H-3→L+2	0.43
	S_0-S_{16}	2.8891 (429.1 nm)	0.0828	H-4→L+1	0.32
				H-4→L+5	0.40
				H-2→L+3	0.44
1a	S_0-S_{13}	2.8659 (432.6 nm)	0.1294	H-5→L+2	0.27
				H-4→L+3	0.26
				H-4→L+5	0.23
				H-3→L	0.21
				H-3→L+4	0.25
				H-2→L+1	0.24
				H-2→L+3	0.22
				H-2→L+5	0.23
	S_0-S_{14}	2.8685 (432.2 nm)	0.1224	H-5→L+2	0.35
				H-4→L+3	0.20
				H-3→L	0.26
				H-3→L+4	0.33
	S_0-S_{15}	2.8781 (430.8 nm)	0.0937	H-5→L	0.30
				H-5→L+4	0.37
				H-3→L+2	0.41
	S_0-S_{16}	2.8792 (430.6 nm)	0.1301	H-4→L+1	0.28
				H-4→L+3	0.27
				H-4→L+5	0.28
				H-2→L+3	0.31
				H-2→L+5	0.27
1b	S_0-S_9	2.7293 (454.3 nm)	0.0133	H-2→L	0.56
				H-2→L+4	0.23

	S_0-S_{13}	2.8697 (432.0 nm)	0.1426	H-5→L+2	0.39
				H-2→L	0.30
				H-2→L+4	0.34
	S_0-S_{14}	2.8731 (431.5 nm)	0.1377	H-4→L+3	0.40
				H-3→L+1	0.30
				H-3→L+5	0.36
	S_0-S_{15}	2.8765 (431.0 nm)	0.1086	H-5→L	0.29
				H-5→L+4	0.31
				H-3→L+3	0.22
				H-2→L+2	0.33
	S_0-S_{16}	2.8781 (430.8 nm)	0.0925	H-5→L+4	0.20
				H-4→L+1	0.29
				H-4→L+5	0.32
				H-3→L+3	0.35
				H-2→L+2	0.21
1c	S_0-S_{13}	2.8630 (433.1 nm)	0.0237	H-4→L+1	0.24
				H-4→L+5	0.31
				H-2→L+1	0.23
				H-2→L+3	0.32
	S_0-S_{14}	2.8637 (433.0 nm)	0.1777	H-5→L	0.24
				H-5→L+2	0.23
				H-5→L+4	0.28
				H-3→L	0.20
				H-3→L+2	0.31
				H-3→L+4	0.20
	S_0-S_{15}	2.8761 (431.1 nm)	0.1700	H-5→L	0.22
				H-5→L+2	0.37
				H-5→L+4	0.23
				H-3→L	0.21
				H-3→L+2	0.26
				H-3→L+4	0.36
	S_0-S_{16}	2.8834 (430.0 nm)	0.1033	H-4→L+1	0.22
				H-4→L+3	0.41
				H-2→L+3	0.23
				H-2→L+5	0.38

[a] Calculated by TDDFT//B3LYP/6-311G(d), based on the DFT//B3LYP/6-311G(d) optimized ground state geometries. [b] Only the low-lying excited states and some allowed transitions were presented. [c] Oscillator strength. [d] Only the main configurations with configuration interaction (CI) coefficients >0.2 are presented. [e] CI coefficients are in absolute values.

Table S2. Selected Electronic Excitation Energies (eV) and Oscillator Strengths (*f*), Configurations of the Low-Lying Triplet Excited States of the complexes **1**[a]

dyes	Electronic transition	Energy (ev)[b]	*f*[c]	Composition	CI[d]
1a	S_0-T_1	2.0958 (591.6 nm)	0.0001	H→L	0.69
1b	S_0-T_1	2.1107 (587.4 nm)	0.0001	H→L	0.69
1c	S_0-T_1	2.0780 (596.7 nm)	0.0001	H→L	0.69
1d	S_0-T_1	2.0929 (592.4 nm)	0.0003	H→L	0.69

[a] Calculated by TDDFT//B3LYP/6-311G(d), based on the DFT//B3LYP/6-311G(d) optimized ground state geometries. [b] Only the selected excited states were presented. [c] Oscillator strength. [d] CI coefficients are in absolute values.