Supporting Information:

Table S1. Hydrogen Bonds for [**1**](ClO_4) [Å and deg]

C–H...O	d(C–H)	d(H...O)	d(C...O)	<(CHO)
C(25)-H(25)...O(1)#1	0.94(5)	2.35(5)	3.235(6)	156(4)
C(2)-H(2)...O(2)#2	0.83(4)	2.57(4)	3.322(7)	152(4)
C(19)-H(19)...O(3)#3	0.87(5)	2.48(5)	3.350(6)	173(4)
C(1)-H(1)...O(4)#2	0.88(4)	2.67(4)	3.405(6)	143(4)

Symmetry transformations used to generate equivalent atoms:

#1 -x,y,-z+1/2 #2 -x+1/2,y+1/2,-z+1/2 #3 x,-y+1,z-1/2

Table S2. Hydrogen Bonds for [**2**]$(ClO_4)_2$ [Å and deg]

C-H...O	d(C-H)	d(H...O)	d(C...O)	<(CHO)
C(20)-H(20)...O(2)#1	0.93	2.61	3.509(12)	163.8
C(19)-H(19)...O(3)#1	0.93	2.68	3.345(11)	129.3
C(4)-H(4)...O(4)#2	0.93	2.48	3.258(10)	141.0
C(13)-H(13)...O(5)#3	0.93	2.46	3.325(12)	155.4
C(24)-H(24)...O(6)#4	0.93	2.35	3.207(11)	153.9
C(18)-H(18)...O(8)#5	0.93	2.46	3.339(11)	157.3
C(9)-H(9)...O(9)#2	0.93	2.41	3.302(13)	160.0

Symmetry transformations used to generate equivalent atoms:

#1 -x,-y+1,-z+1 #2 -x,-y+1,-z #3 -x+1,-y+1,-z

#4 -x-1,-y+1,-z+1 #5 -x,-y,-z+1

Table S3. Hydrogen Bonds for [**3**](ClO_4) [Å and deg]

C-H...O	d(C-H)	d(H...O)	d(C...O)	<(CHO)
C(10)-H(10)...O(3)#1	0.93	2.44	3.272(7)	149.0
C(8)-H(8)...O(6)#2	0.93	2.66	3.205(11)	118.5
C(23)-H(23)...O(5)#3	0.93	2.40	3.137(9)	135.7

Symmetry transformations used to generate equivalent atoms:

#1 -x+1,-y,-z+1 #2 -x+2,-y,-z+1 #3 -x+1,-y,-z

Figure S1. Intermolecular hydrogen bonding in [**1**](ClO_4).

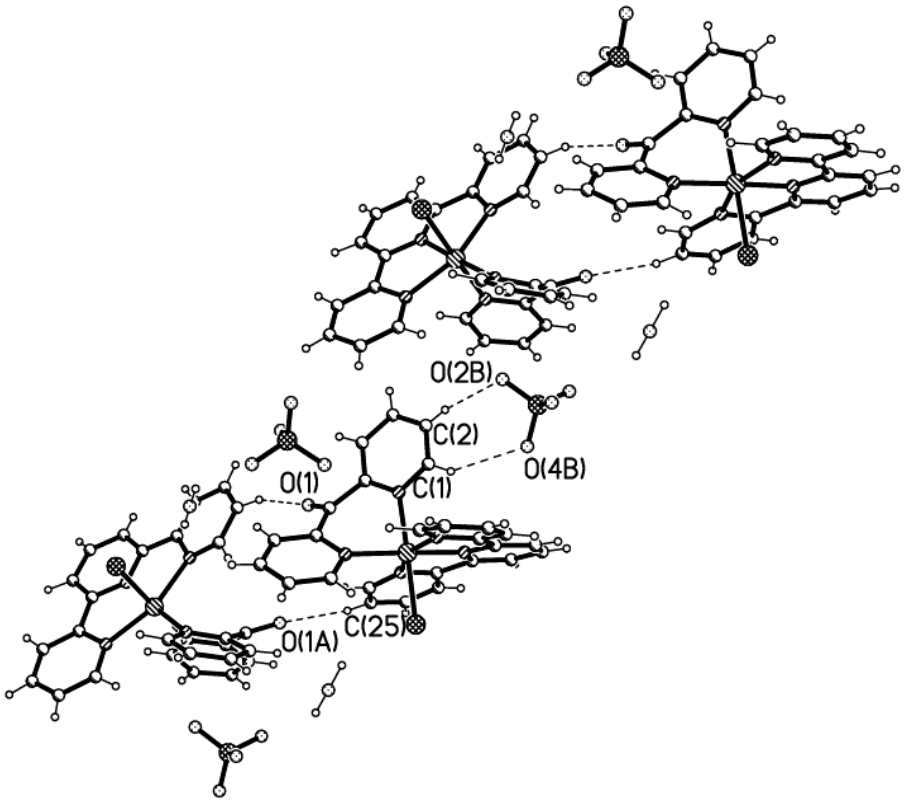

Figure S2. Intermolecular hydrogen bonding in [**2**]$(ClO_4)_2$.

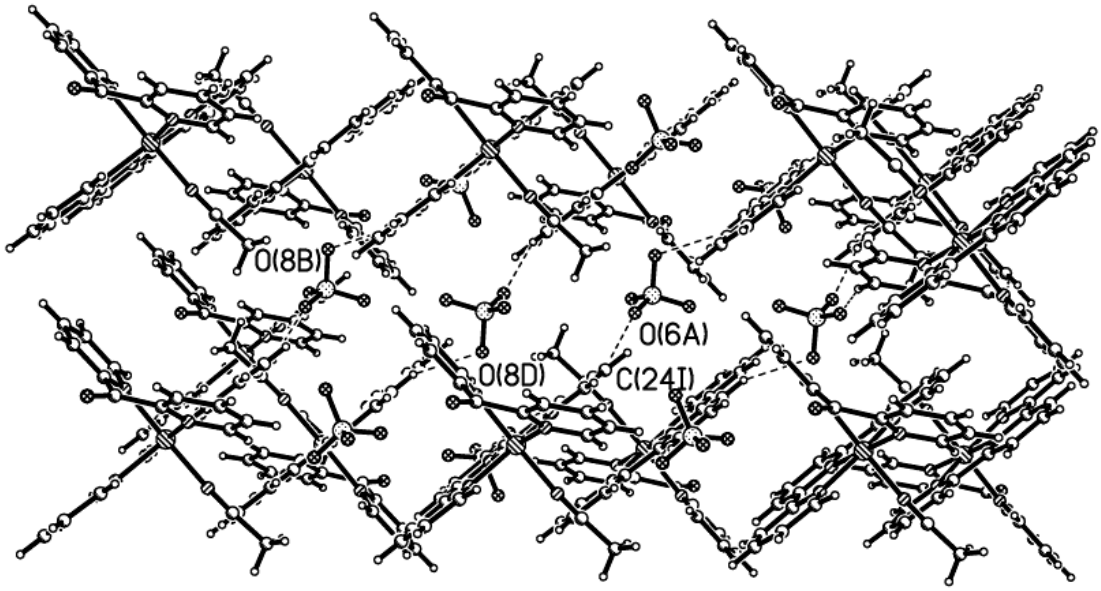

Figure S3. Intermolecular hydrogen bonding in [**3**](ClO_4).

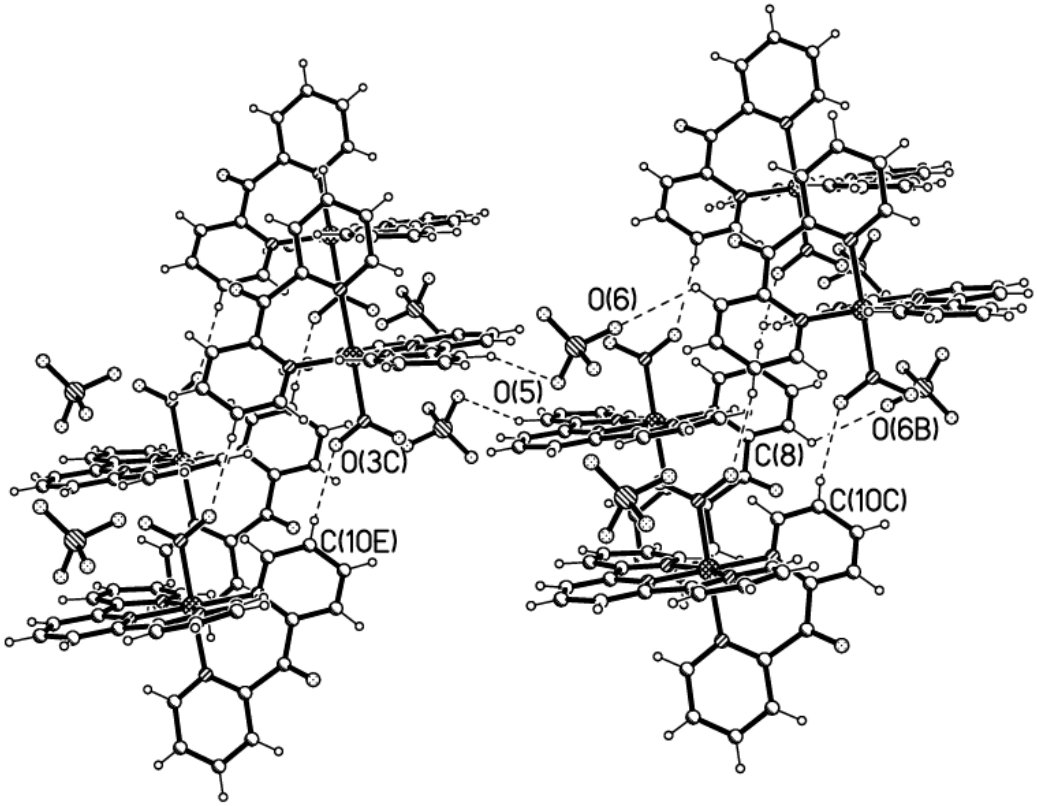

Figure S4. ^{1}H NMR spectra of (a) [**1**](ClO_4) in $(CD_3)_2SO$, (b) [**2**](ClO_4)$_2$ in D_2O and (c) [**3**](ClO_4) in $(CD_3)_2SO$.

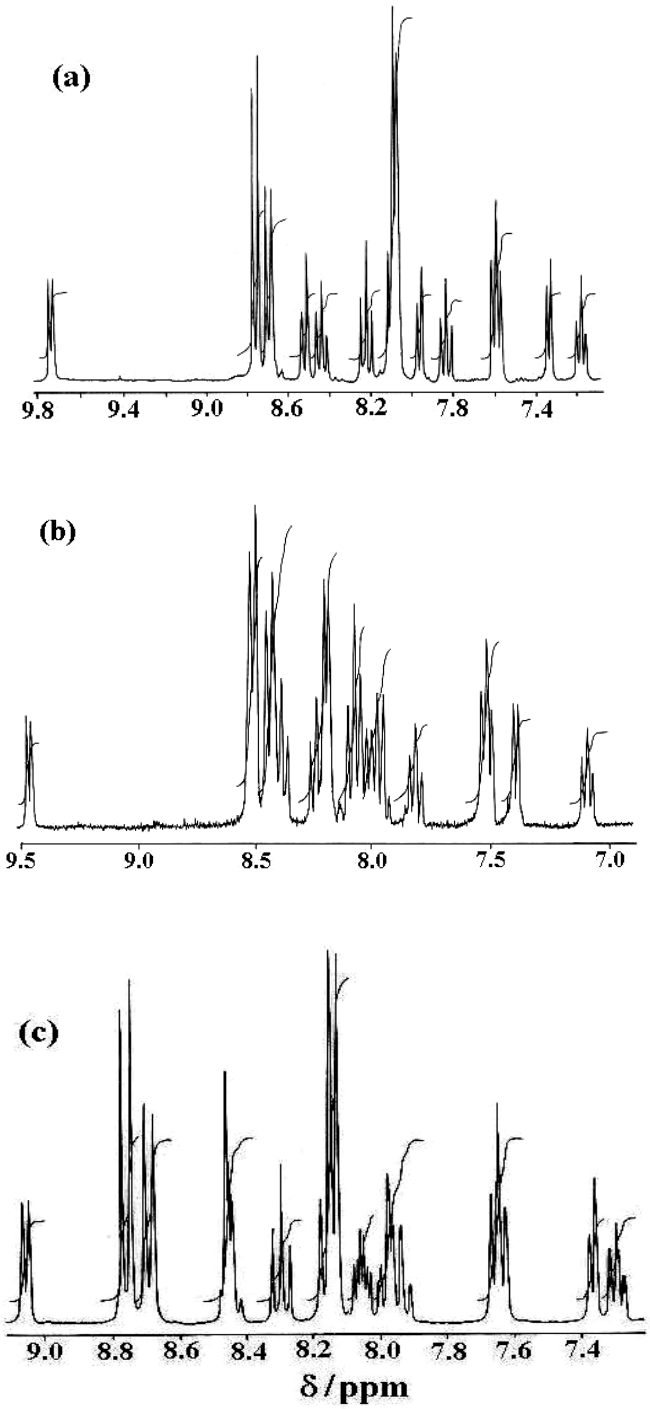

Figure S5. IR spectrum of [**4**]$(ClO_4)_3$ in KBr disk.

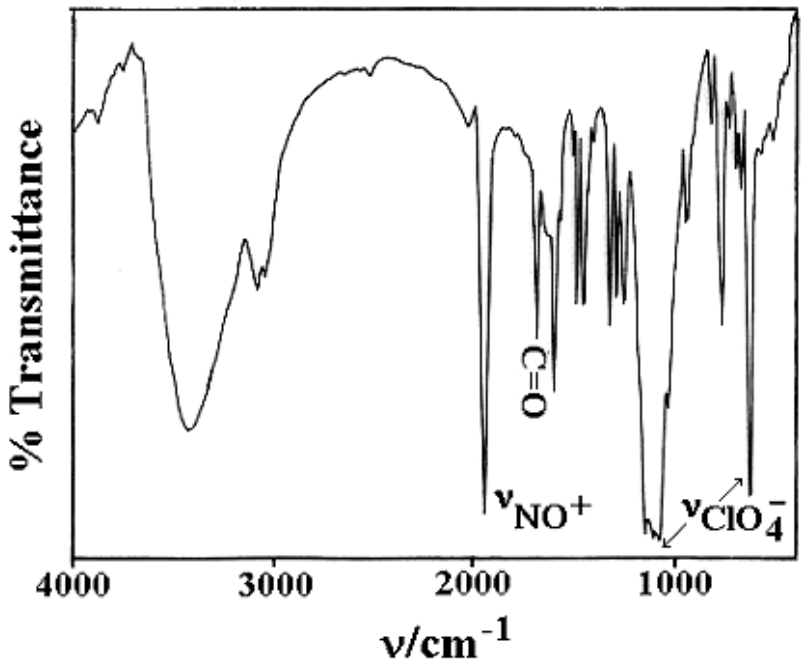

Figure S6. Time evolution of the electronic spectra of a changing solution $[Ru^{II}(trpy)(dpk)(NO^{+})]^{3+} \rightarrow [Ru^{II}(trpy)(dpk)(NO_2)]^{+}$ in acetonitrile–water (10:1) at 308 K. The arrows indicate increase or decrease in band intensities as the reaction proceeds.

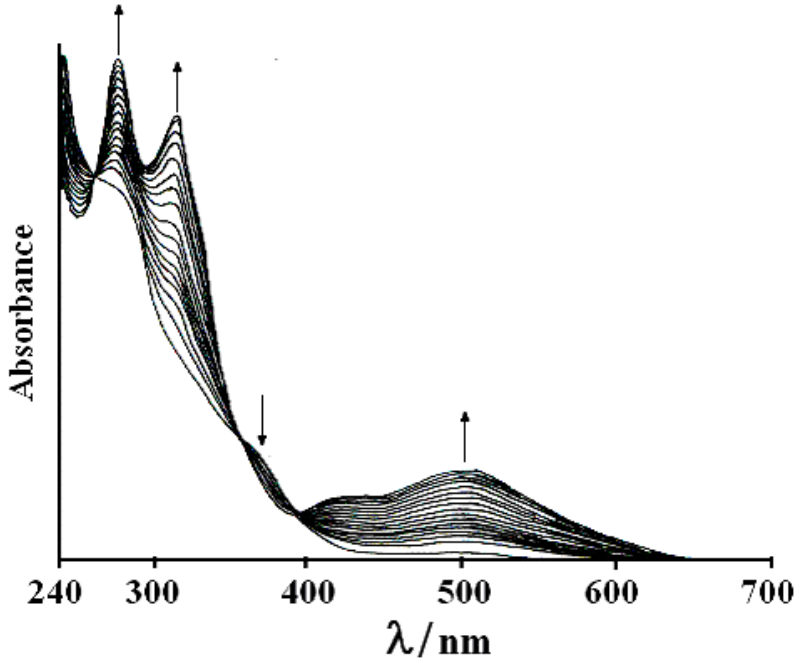

Figure S7. Electronic spectra in acetonitrile of [**1**](ClO_4), [**2**]($ClO_4)_2$, [**3**](ClO_4) and [**4**]($ClO_4)_3$.

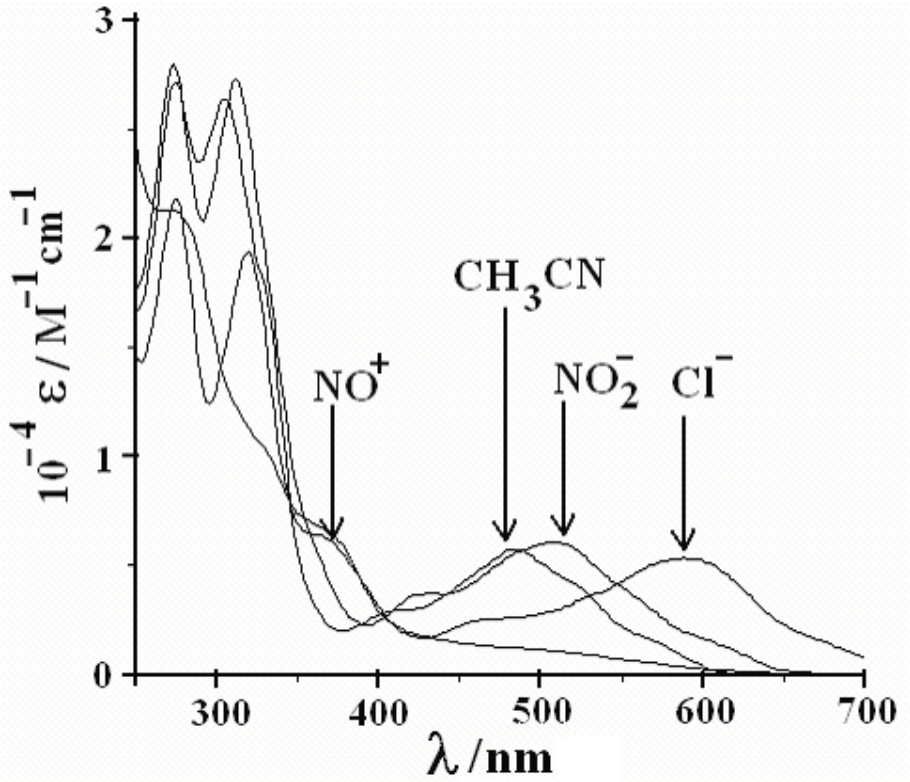

Figure S8. Emission spectra of [**1**](ClO_4), [**2**](ClO_4)$_2$ and [**3**](ClO_4) in 4:1 EtOH–MeOH at 77 K.