Electronic Supplementary Material (ESI) for RSC Advances.

Supplementary Data

(Figures S1-S2 and Tables S1).

Table S1 HOMO and LUMO energy values (in eV) calculated by DFT method

Parameters	Compound	B3LYP/LanL2DZ
E_{total} (hartree)	**1**	-1095
	2	-1367
HOMO energy (eV)	**1**	-5.42 (α-spin, SOMO)
	2	-5.55 (α-spin, SOMO)
LUMO energy (eV)	**1**	-1.77
	2	-1.88
$\Delta E_{HOMO-LUMO}$ (eV)	**1**	3.65
	2	3.67

1 hartree = 627.5095 kcal/mol = 27.2116 eV

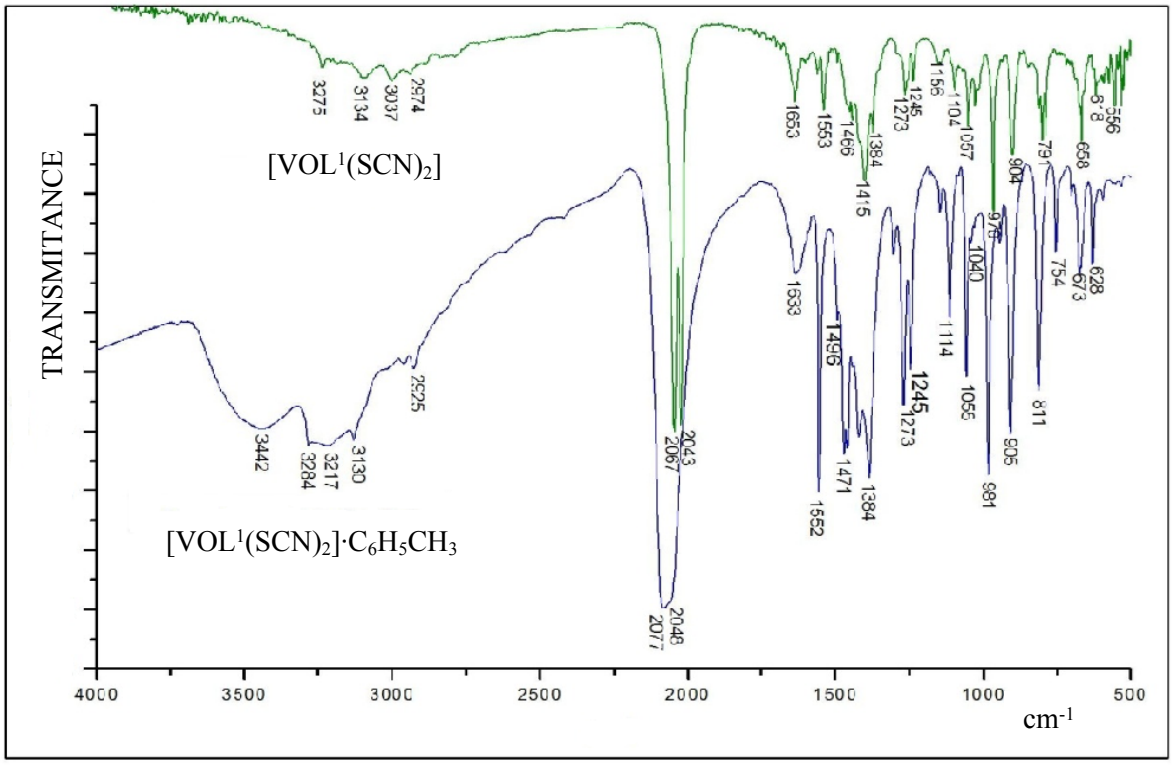

Fig S1 . Experimental infrared absorption spectra of [$VOL^1(SCN)_2$] (**1**) and $VOL^1(SCN)_2]\cdot C_6H_5CH_3$ (**2**).

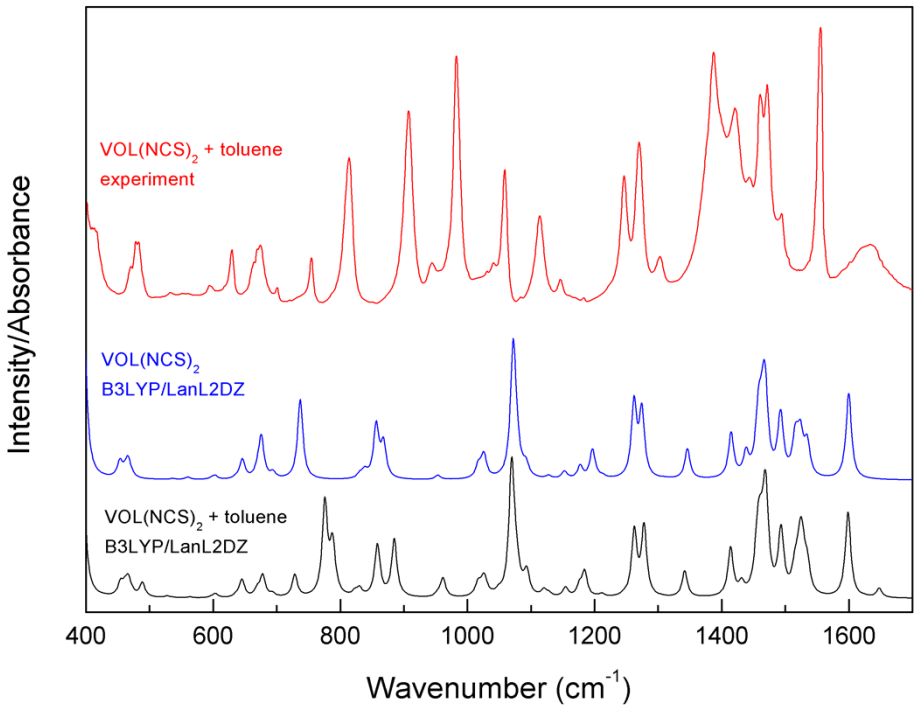

Fig. S2 The calculated infrared absorption spectra of **1** (blue) and **2** (black) together with the experimental spectrum of **2** (red). The spectra were vertically offset for clarity.

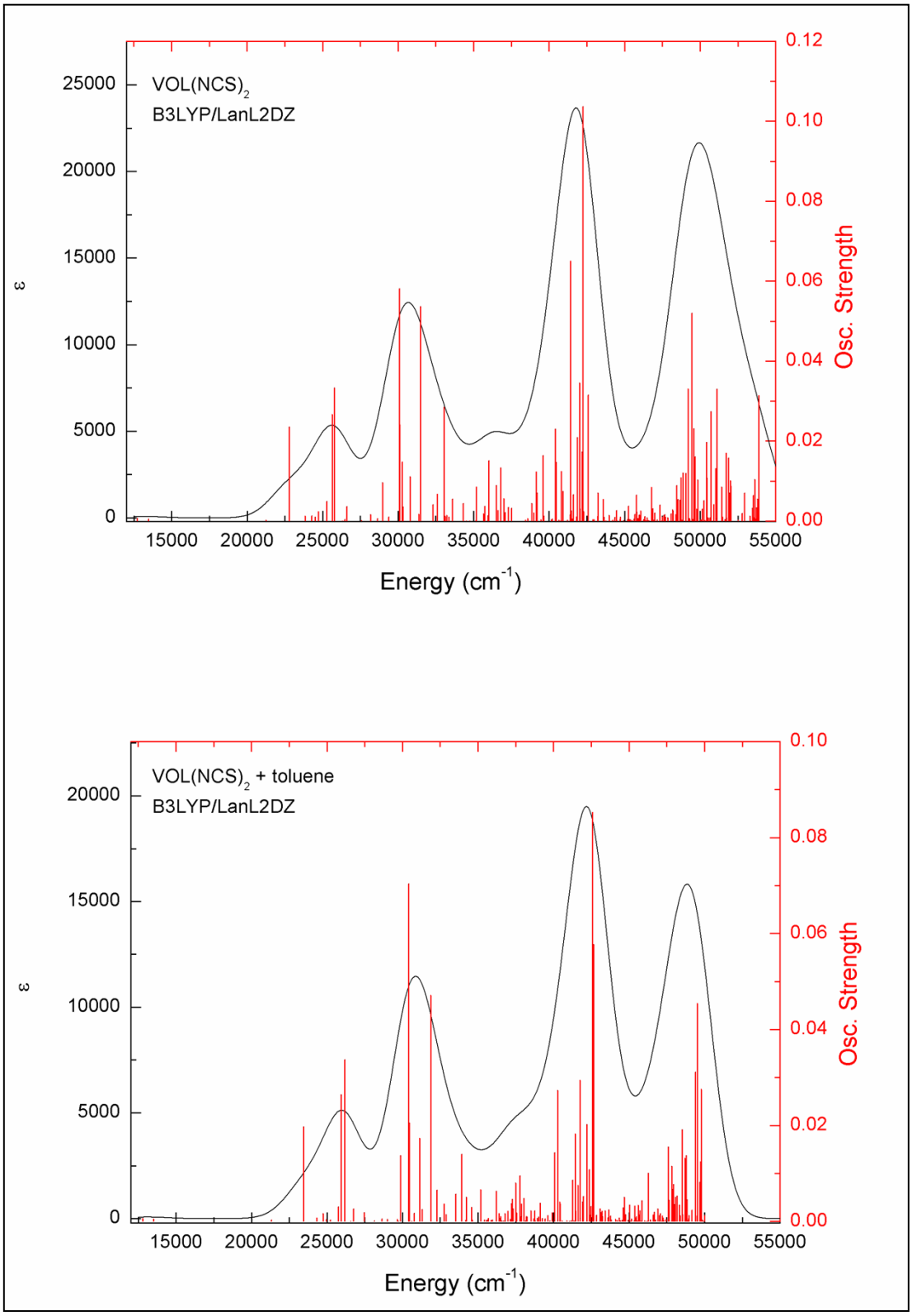

Fig. S3. Calculated transitions (red colour, right scale) with the convoluted electronic absorption spectrum (black colour, left scale) of **1** and **2**. Theory level: B3LYP/LanL2DZ.