Supporting Information for

In situ Formation of New Organic Ligands to Construct Two Novel Self-charge-transfer Pb(II)-based Frameworks

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Materials and methods

All the chemicals used for synthesis are of analytical grade and commercially available. $Pb(SC_6H_5)_2$ was synthesized according to the reported method.[1] The IR spectrum was obtained on a Perkin-Elmer FT-IR spectrophotometer in the 400–4000 cm^{-1} region with a KBr pellet. Elemental analyses were performed on a Perkin-Elmer 2400 Elemental Analyzer. Thermal stability studies were carried out on a TGA Q500 instrument under N_2 at a heating rate of 10 °C. The DC magnetic susceptibility measurements were made on an MPMS magnetometer at temperatures between 5.0 and 273 K. The optical diffuse reflectance spectra were measured at room temperature on a Perkin-Elmer Lambda 900 UV–vis-NIR spectro-meter equipped with an integrating sphere. $BaSO_4$ was used as the reference material, and the polycrystalline samples were ground well before the measurement. The absorption (α/S) data were calculated from the reflectance using the Kubelka–Munk function: $\alpha/S = (1-R)^2/2R$, in which R is the reflectance at a given wavelength, α is the absorption coefficient, and S is the scattering coefficient.[2] Powder X-ray diffraction data were recorded on a Bruker D8 Advance diffractometer with a graphite-monochro-matized Cu *Kα* radiation. The operating 2θ angle ranges from 10° to 55°. Simulation of the XRD pattern was carried out by the single-crystal data and diffraction-crystal

module of the Mercury (Hg) program version 1.4.2 available free of charge via the Internet at http://www.iucr.org.

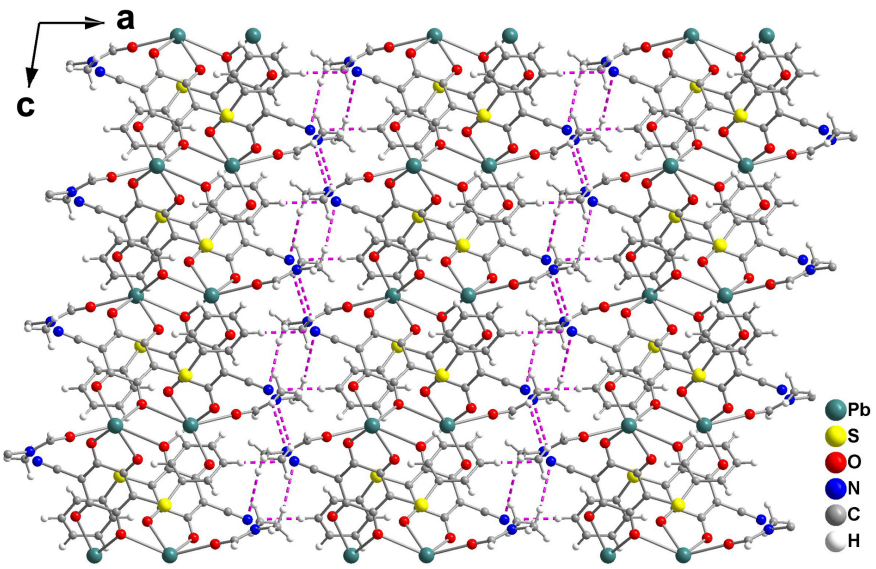

Figure S1. View of the 2D layered structure of **2** along the *b*-axis.

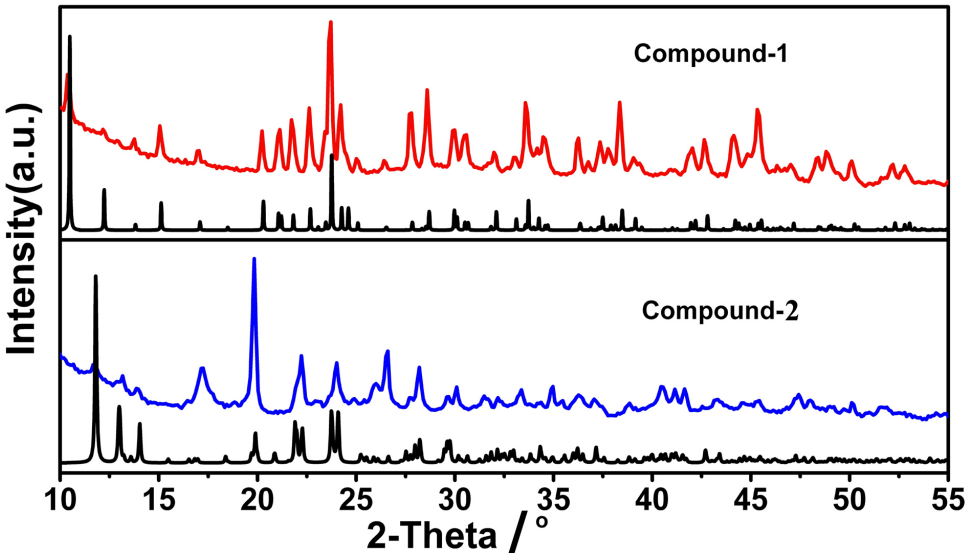

Figure S2. Experimental and simulated XRD patterns of compounds **1** and **2**.

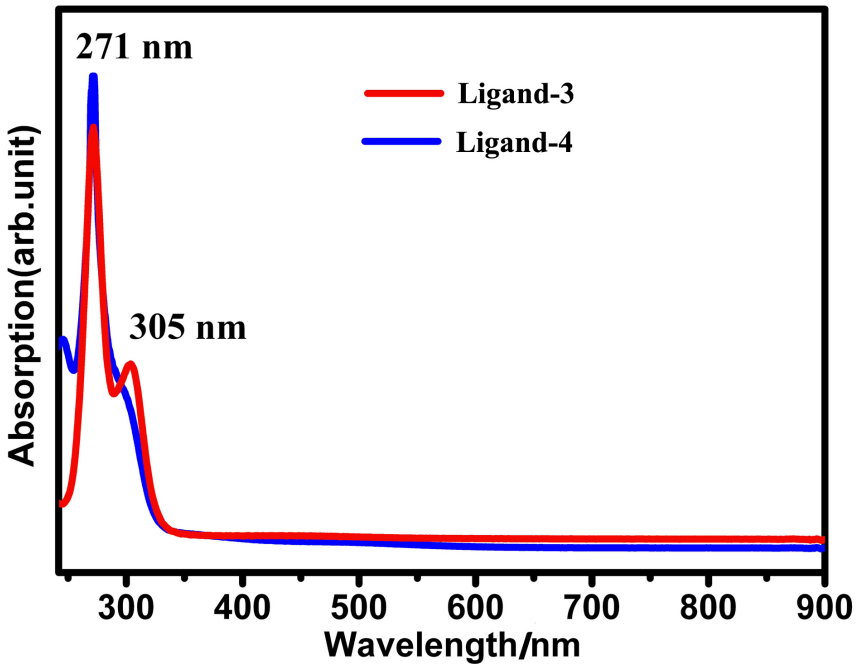

Figure S3. The UV-Vis absorption of in-situ formed organic ligands **3** and **4**.

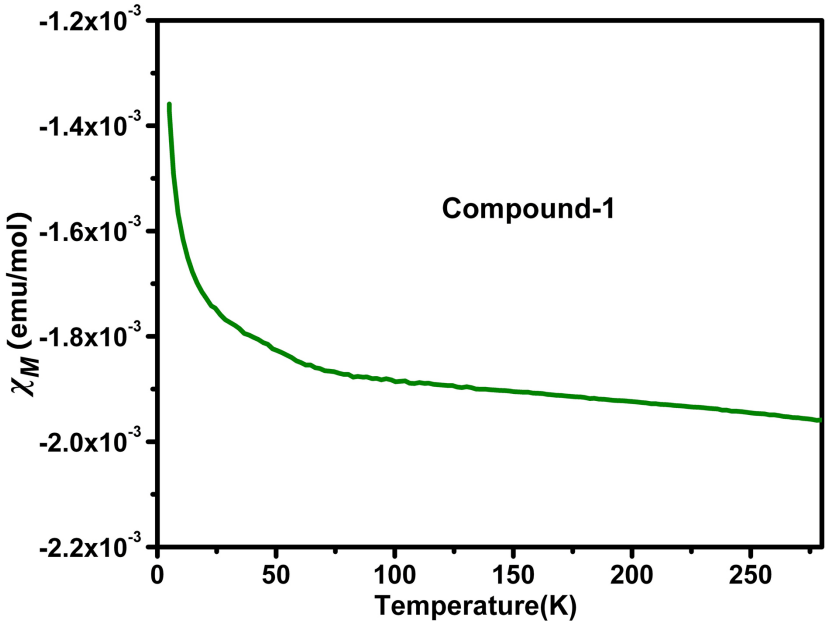

Figure S4. Molar susceptibilities as a function of temperature for **1**, measured in an applied field of 5 kOe.

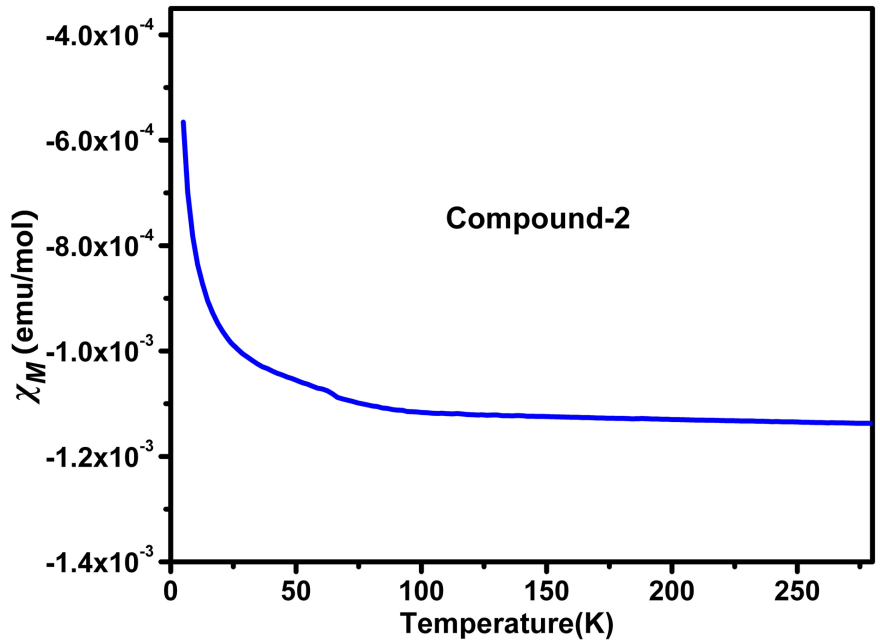

Figure S5. Molar susceptibilities as a function of temperature for **2**, measured in an applied field of 5 kOe.

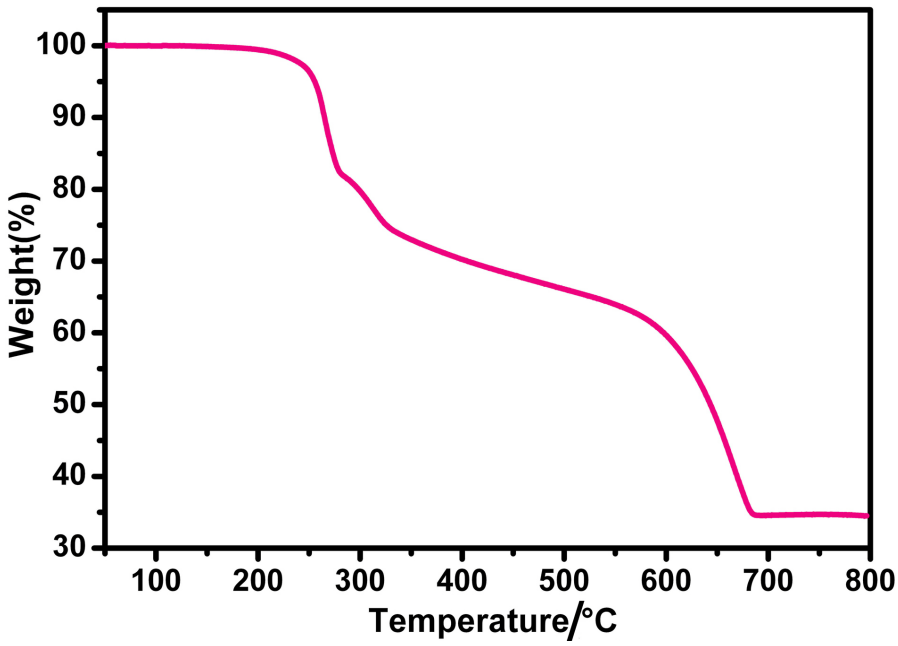

Figure S6. TGA curve of **1** in N_2 with a heating rate of 10 °C.

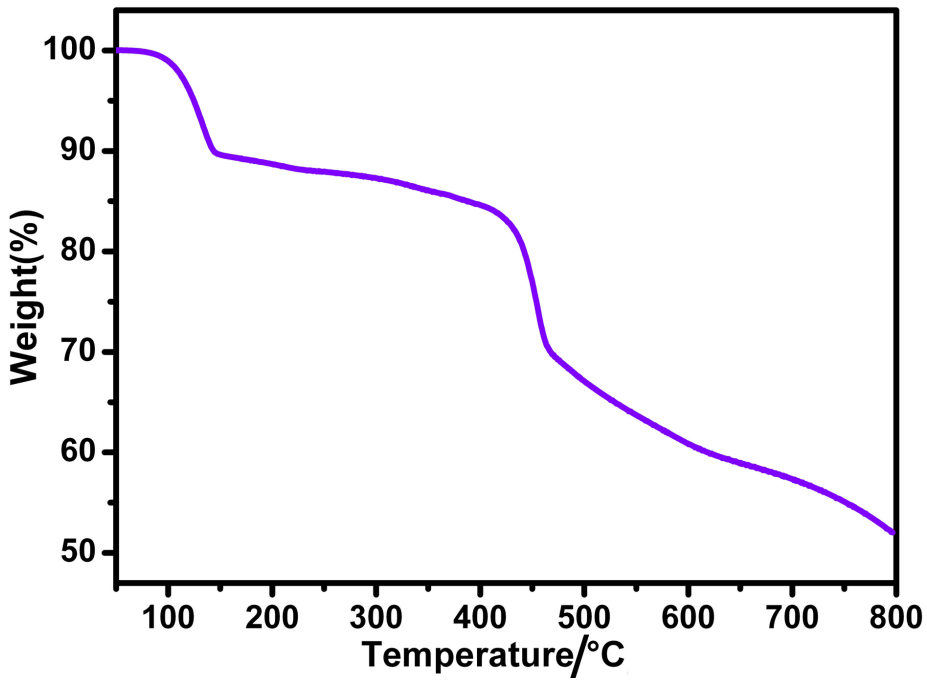

Figure S7. TGA curve of **2** in N_2 with a heating rate of 10 °C.

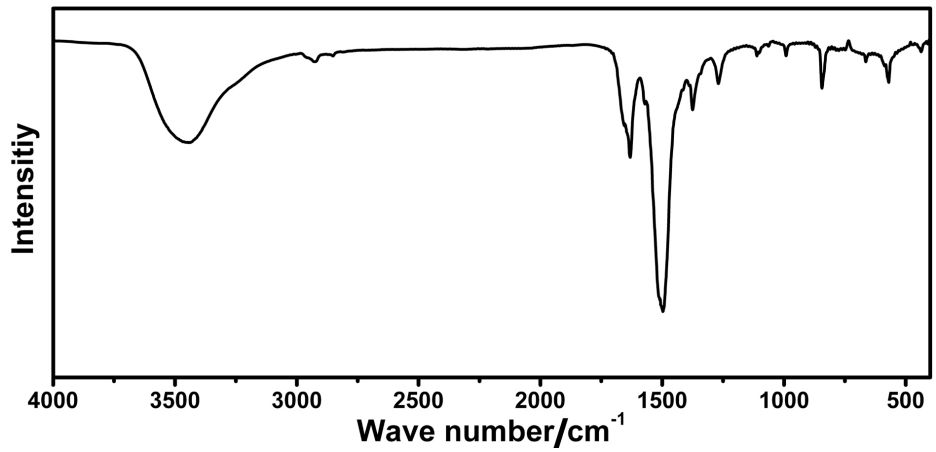

Figure S8. FT-IR spectrum of **1** (cm^{-1}): 2955 (w, ν(C–H)), 1631 (s, δ(C=O)), 1497 (vs, δ (C=C)), 1375 (m, ν(C–C)), 1269 (m, ν(C–N)), 1111 (w, ν(C–N)), 991 (w, ν(C–O)), 844 (m, ν(C–O)), 570 (m ν(C–Cl))

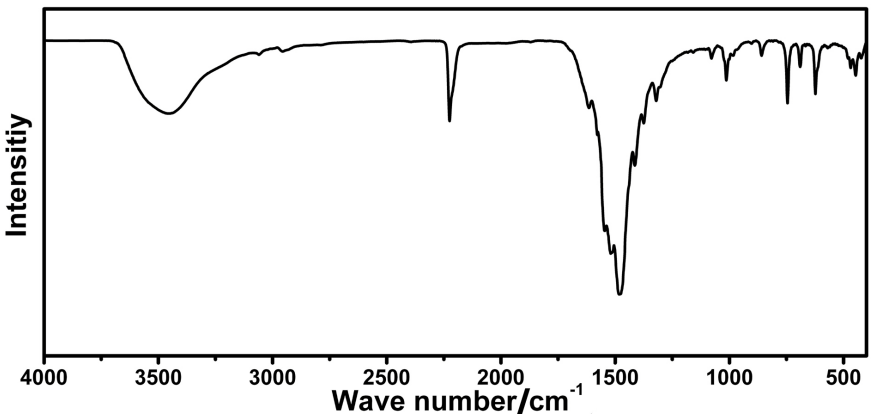

Figure S9. IR spectrum of **2** (cm^{-1}): 2929 (w, ν(C–H)), 2225 (m, ν(C≡N)), 1614 (m, δ(C=O)), 1578 (s, δ(C=C)), 1545 (vs, δ(C=C)), 1519 (vs, δ(C=C)), 1482 (vs, δ (C=C)), 1413 (s, δ(C=C)), 1374 (m, ν(C–C)), 1320 (m, ν(C–N)), 1078 (w, ν(C–S)), 1013 (w, ν(C–S)), 859 (w, δ(=CH)), 745 (m, δ(=CH)), 623 (m δ(=CH))

References

1 R. A. Shaw and M. Woods, *J. Chem. Soc.,* **1971**, 1569.

2 G. Kortüm, *Reflectance Spectroscopy*, Springer, New York, **1969**.