

# Supporting Information

## The Power of Three: Incremental Increase in the Ligand Field Strength of N-Alkylated 2,2'-Biimidazoles Leads to Spin Crossover in Homoleptic Fe(II) Complexes

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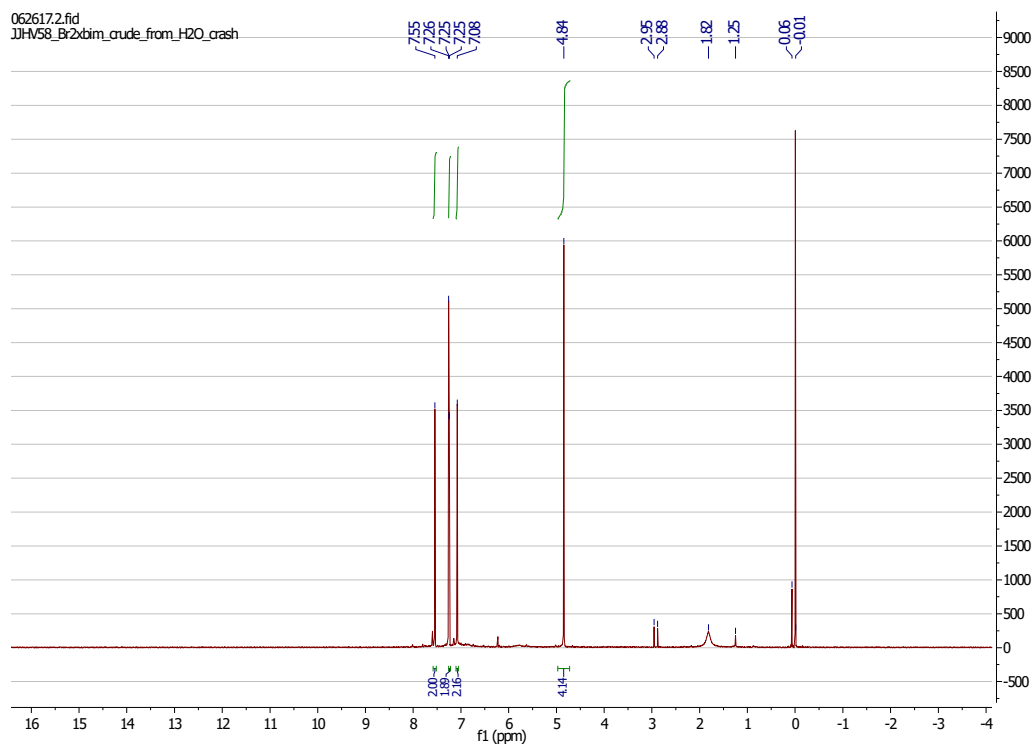


Figure S1.  $^1\text{H}$ -NMR spectrum of  $\text{L}_2$

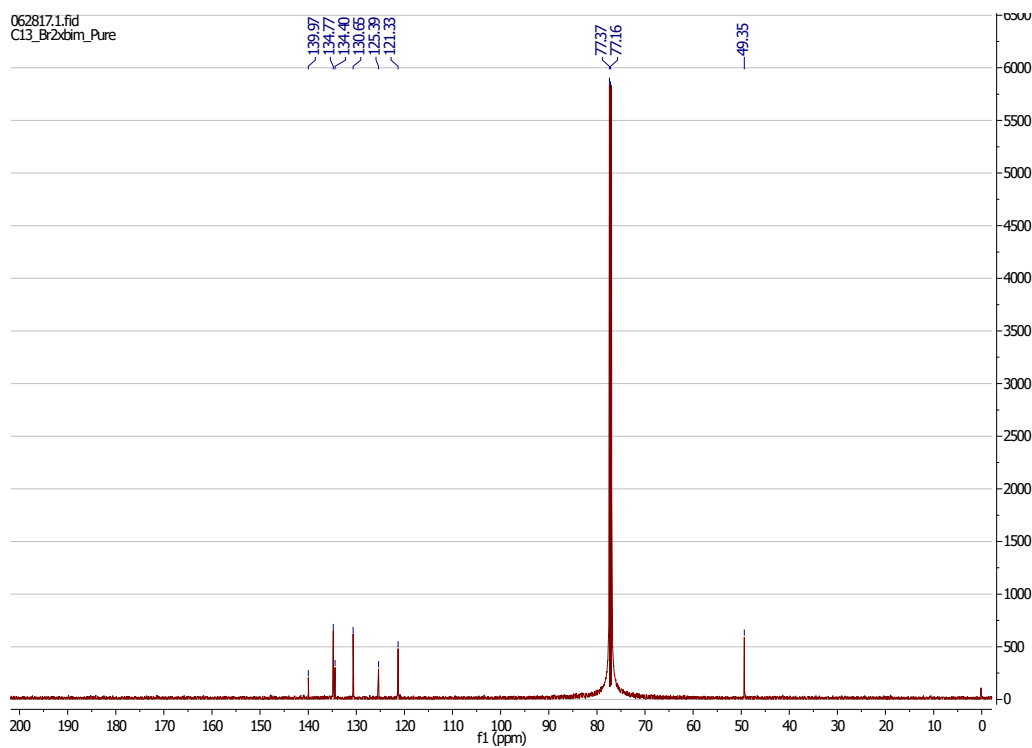


Figure S2.  $^{13}\text{C}$ -NMR spectrum of  $\text{L}_2$

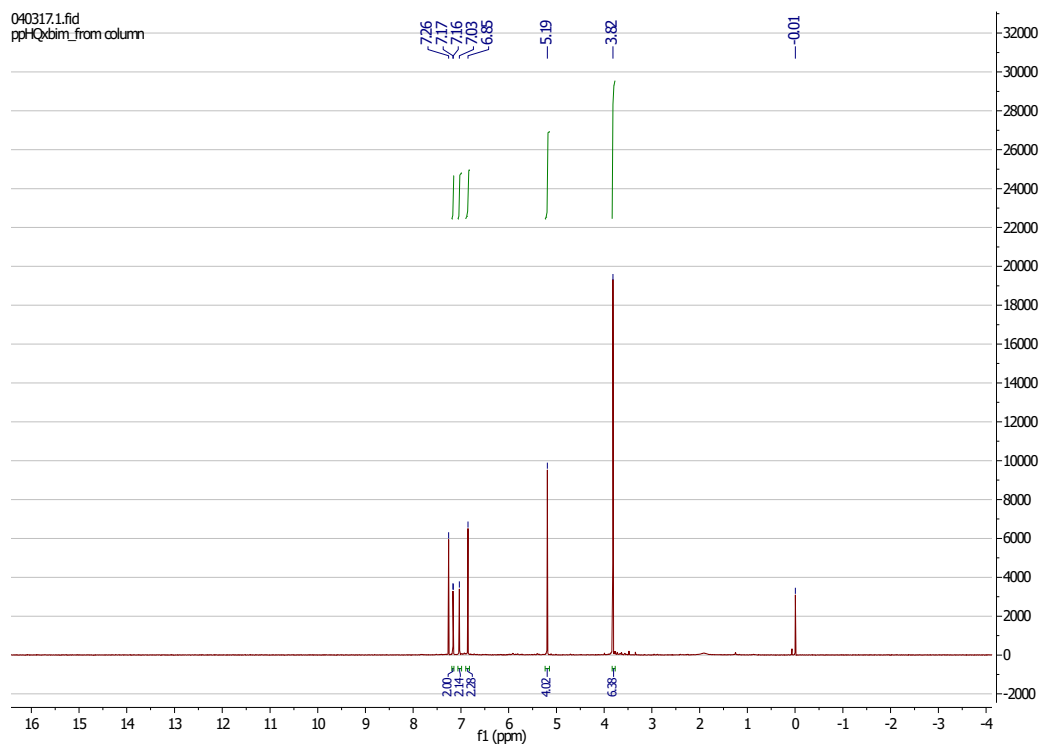


Figure S3.  $^1\text{H}$ -NMR spectrum of  $\text{L}_3$

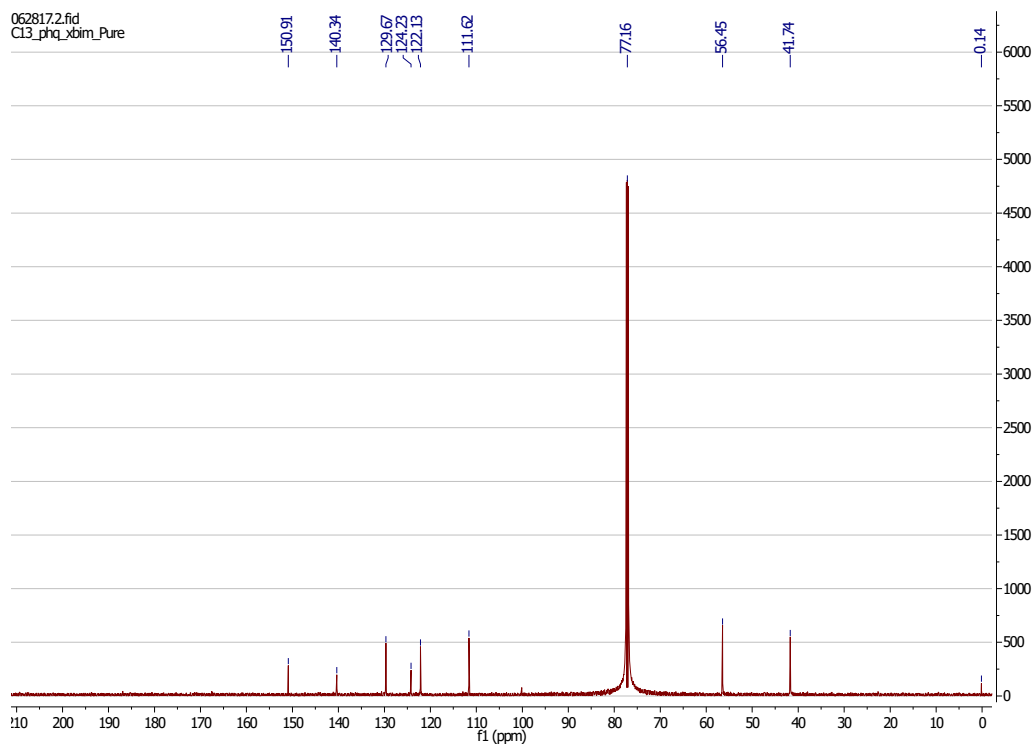


Figure S4.  $^{13}\text{C}$ -NMR spectrum of  $\text{L}_3$

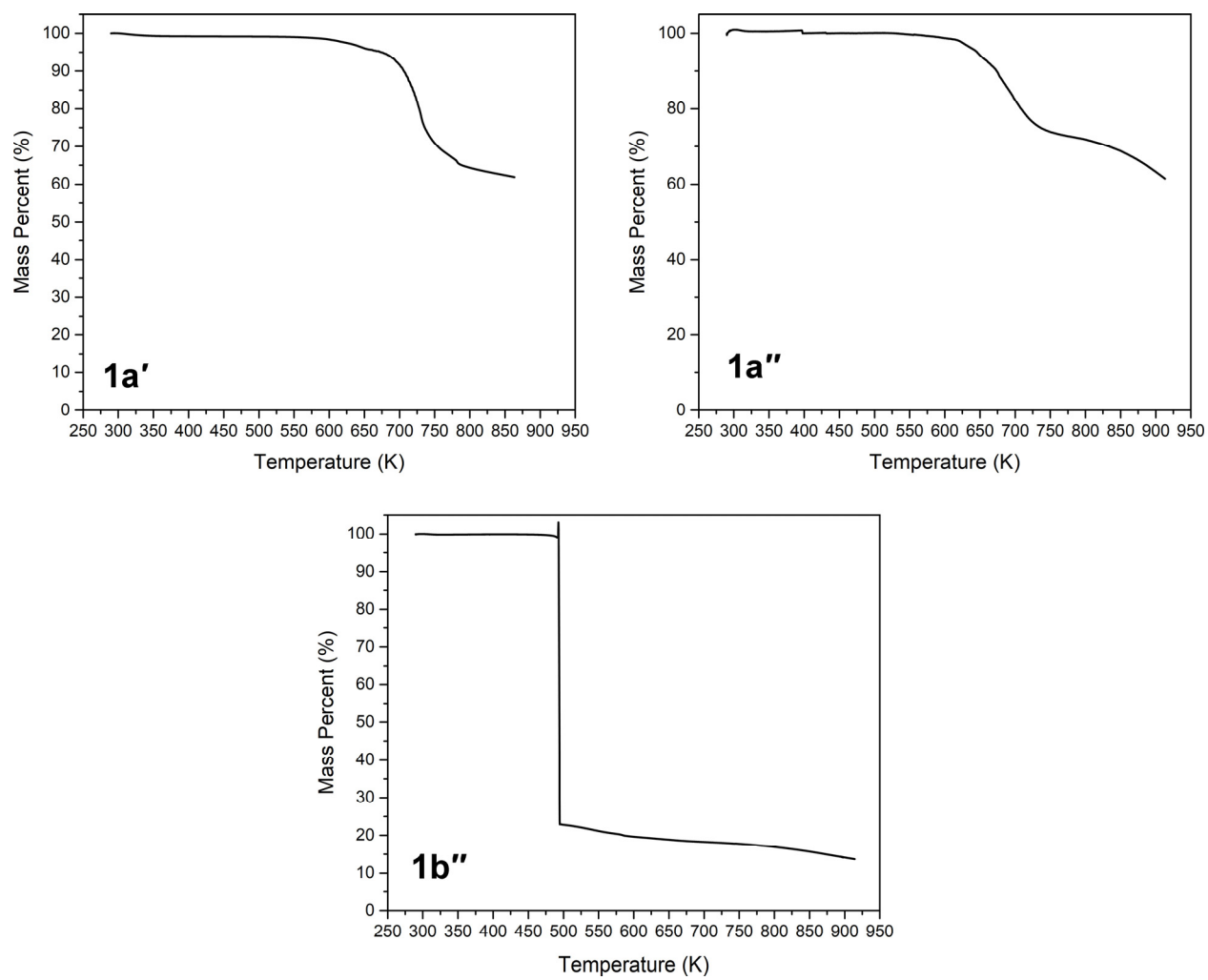


Figure S5. TGA plots of **1a'**, **1a''**, and **1b''** measured at a heating rate of 10 K/min.

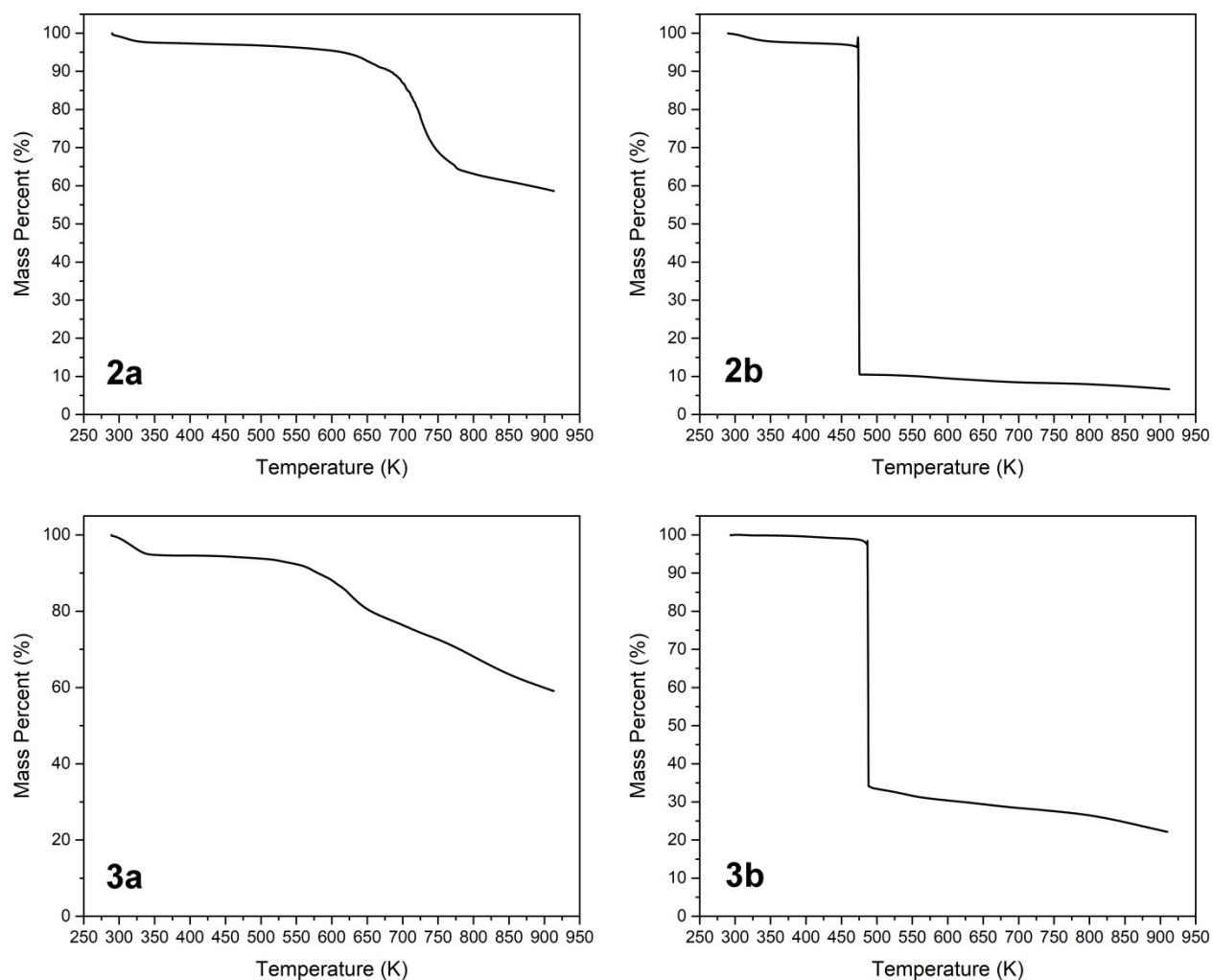


Figure S6. TGA plots of **2a**, **2b**, **3a**, and **3b** measured at a heating rate of 10 K/min.