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## Spin Crossover in Iron(II) Imidazolylmethylene-aryl amine Complexes: Tuning by the substituent group on the ligand

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The new ligands imidazol-4-ylmethylene-aryl amine (4-imaAr; Ar = Bp, Fl) have been prepared by reaction of the aryl amine with imidazole-4-carboxaldehyde.[1,2] Reaction of the ligands with  $[\text{Fe}(\text{H}_2\text{O})_6][\text{ClO}_4]_2$  in MeOH or EtOH yields the complexes,  $[\text{Fe}(4\text{-imaBp})_3][\text{ClO}_4]_2 \cdot \text{EtOH}$  1,  $[\text{Fe}(4\text{-imaBp})_3][\text{ClO}_4]_2 \cdot 2\text{MeOH}$  2,  $[\text{Fe}(4\text{-imaFl})_3][\text{ClO}_4]_2$  3, and  $[\text{Fe}(4\text{-imaFl})_3][\text{ClO}_4]_2 \cdot 3\text{H}_2\text{O}$  4. X-ray crystallographic studies reveal strong C-H $\cdots\pi$  and hydrogen bonding interactions or  $\pi$ - $\pi$  interactions leading to high cooperativity. SQUID magnetic susceptibility studies show that spin crossover varies from gradual to abrupt with the spin transition occurring at temperatures from ca. 150 to 400 K. The substituent groups on the 4-ima ligand significantly impact the spin crossover characteristics.

### Keywords

spin crossover, iron(II) complexes, imidazolyl diimine ligands, molecular magnetism

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