

# A LOW-COORDINATE N<sub>2</sub>-BRIDGED DICOBALT(I) COMPLEX WITH NON-AUFBAU ELECTRONIC GROUND STATE

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Binding and activation of dinitrogen at metal ions is a key step in the fixation of this inert substrate into bioavailable N-containing compounds, most prominently in the industrial Haber-Bosch process and in the enzymatic NH<sub>3</sub> formation mediated by nitrogenase. In the latter, N<sub>2</sub> is reduced at low-coordinate high-spin iron ions at the central region of the oligometallic FeMo cofactor. Numerous synthetic complexes featuring coordinated N<sub>2</sub> have been reported with the aim of reductive N<sub>2</sub> functionalization,[1,2] and in this field cobalt is emerging as a particularly promising metal ion of the 3d series [3-5], after iron. However, low-coordinate high-spin cobalt/N<sub>2</sub> complexes remain rare.[3] In this work, we present a low-coordinate high-spin dicobalt/N<sub>2</sub> complex,  $\{[(\text{IAd})\text{Co}(\text{C}(\text{TMS})_3)]_2(\mu\text{-N}_2)\}$  (**2**), formed from the putative two-coordinate cobalt(I) complex  $[(\text{IAd})\text{Co}(\text{C}(\text{TMS})_3)]$  (**1**) featuring a trigonal planar geometry of the cobalt center, which is supported with a bulky N-heterocyclic carbene (NHC) ligand IAd and a bulky carbanion group. The structural and spectroscopic characterization of **2** as well as its N<sub>2</sub> binding equilibrium in solution[6] will be discussed. Synchrotron-based K-edge XAS experiments accompanied by DFT and CASSCF/NEVPT2 calculations were performed to understand the rare non-aufbau electronic structure. Furthermore, **2** was revealed to possess unexpected SMM property and catalytic reactivity.

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