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Reversible mechanism for spin crossover in transition-metal cyanides MUKUL KABIR, KRYSTYN J. VAN VLIET, Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA — Spin transitions generally occur in compounds of octahedrally coordinated $3d$ transition metal ions. These transitions can be induced by external perturbations such as light, heat, pressure, magnetic field, and chemical substitution. Transition metal cyanides are one such material, which exhibit *reversible* spin transition while perturbed with light at T <10 K. Here we report the first-principles (DFT+U) study of anhydrated $KCoFe(CN)_6$. We find that the complete spin transition from the low spin ground sate ($S = 0$) to a high spin ($S = 2$) state takes place due to intra-atomic and inter-atomic charge transfers in two steps. In the first step a d-electron is transferred from Fe to Co through cyanide ligand, which is followed by the d-electron rearrangement in the Co. This spin transition is strongly correlated with the internal lattice, and we find as large as 10% extension of the Co−N bond via a Jahn-Teller active (tetragonally distorted) lattice in the intermediate spin ($S = 1$) state. The calculated energy required for this transition is in agreement with experiments. We further predict that this spin transition in such materials can be induced, and further tuned, by external pressure to enable realization of such reversible transitions at ambient temperatures.

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