Exploring Spin-Crossover Materials with Machine Learning

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The observed transitions in spin-switching materials, and their associated bistable behavior, arise from a complex combination of both short- and long-range interactions that also are coupled with molecular properties. Those, in turn, are sensitive to local chemical environments [Coord. Chem. Rev. **435**, 213819 (2021)]. Together, these give rise to different possible thermally induced transition curves. The spin conversion relies on a delicate balance between the adiabatic crossover energy, vibrational free energy, and magnetic entropy [C. R. Chimie **21**, 1060 (2018)]. Typically, these crossover energies are no larger than about 10 kJ/mol, a magnitude that challenges state-of-the-art density functional approximations and requires methodologies nearing the limit of "chemical accuracy" [Perdew and Schmidt in *Density Functional Theory and Its Application to Materials*, edited by V. Van Doren, C. Van. Alsenov, and P. Geerlings (AIP, Melville, NY, 2001)]. This poses a particular challenge in the context of high-throughput screening because of the large number of atoms in voluminous ligands and the sensitivity of the spin-conversion energy to diverse computational choices [Comput. Mater. Sci. **206**, 111161 (2022)].

This work explores an alternative to conventional screening approaches based on equivariant graph neural networks [arXiv:2102.09844], trained on a set of 1592 metal-organic materials. The architecture exploits the symmetry group of the unit cell to produce representations useful for predicting molecular and solid-state properties. For that purpose, we compare two strategies, namely, use of supervised learning, and a combination of unsupervised followed by supervised learning in a two-step implementation. We compare results for both approaches and discuss their potential as a tool for screening spin-crossover compounds.

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