Can simple exchange heuristics guide us in the machine learning of magnetic properties of solids?

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Environmental and scarcity issues of common functional magnetic materials for, e.g., permanent magnets have intensified the search for rare-earth-free alternatives. This challenge is increasingly met by machine learning of magnetic properties of transition-metal compounds.
Surprisingly, bond-angle-derived features were not found to be relevant for magnetic structure prediction in previous studies using DFT-computed labels.[1, 2] This contrasts with the Kanamori-Goodenough-Anderson (KGA) rules of superexchange, present in every magnetism textbook.
These semiempirical rules predict whether a nearest-neighbor magnetic interaction in insulators is FM or AFM based on the bond angle, orbital symmetry, and orbital occupancy.[3, 4] For some cases, the rules can be simplified further to only consider the bond angle of neighboring magnetic sites (KGA rules of thumb).
We review magnetism—bond angle trends within the MAGNDATA database,[5] the largest collection of experimentally determined magnetic structures. Observed trends follow the KGA rules of thumb, and exceptions can be rationalized. In contrast, bond angles in a popular theoretical DFT database[1] show very different trends and do not depend on the magnetic ordering.
Building on our analysis, we engineer heuristic-derived features for the machine learning of magnetic structures. We introduce a new, informative label for predicting magnetic structures that can be extended to magnetic sites and structures of arbitrary complexity. We show that features derived from the heuristic are of high importance for this machine learning task. Beyond this, our model enables the prediction of non-collinear magnetic structures. Further, we analyze local and global structural trends of non-collinear magnets in the MAGNDATA database.

References

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