Learning Magnetic Exchange Coupling in Chemically Functionalized Open–Shell Nanographenes

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Open-shell nanographenes offer a versatile platform for investigating how chemical design controls magnetic interactions. In dimers of [3]triangulene units, spin moments predominantly localized on the two fragments interact through an exchange coupling constant J, whose magnitude and sign depend on both chemical functionalization and molecular geometry. Understanding these relationships is essential for guiding the design of molecular magnetic systems.

We developed a high–throughput computational workflow to generate and investigate a library of 1000 chemically functionalized [3]triangulene dimers, providing a dataset for machine learning prediction of their exchange coupling. The magnetic exchange couplings are computed using DFT calculations and span a range of approximately 6 meV.

Analysis of the computed electronic and magnetic properties revealed complex structure–property relationships that could not be readily rationalized using simple structural descriptors.

To address this challenge, we integrated the Smooth Overlap of Atomic Positions (SOAP) representation [1] and the Regularized Entropy Match (REMatch) kernel [2] with Gaussian Process Regression [3] and the Sketch-map visualization of chemical space [4]. On an independent test set, the model achieved a coefficient of determination of approximately $R^2 = 0.80$ and a mean absolute error of 0.48 meV.

These results demonstrate the potential of similarity–based machine learning to capture the dependence of magnetic exchange on molecular structure for this family of open-shell nanographenes. The workflow enables rapid exploration of the investigated chemical space and provides a basis for prioritizing candidate structures for further electronic-structure calculations.

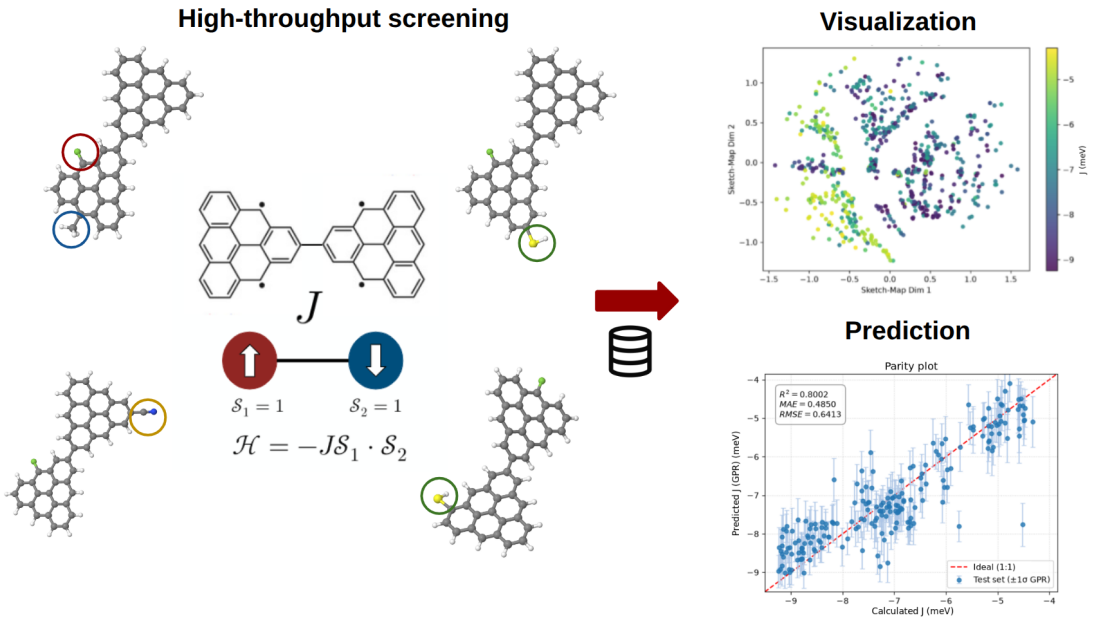

Figure 1: Schematic representation of the computational workflow.

References

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