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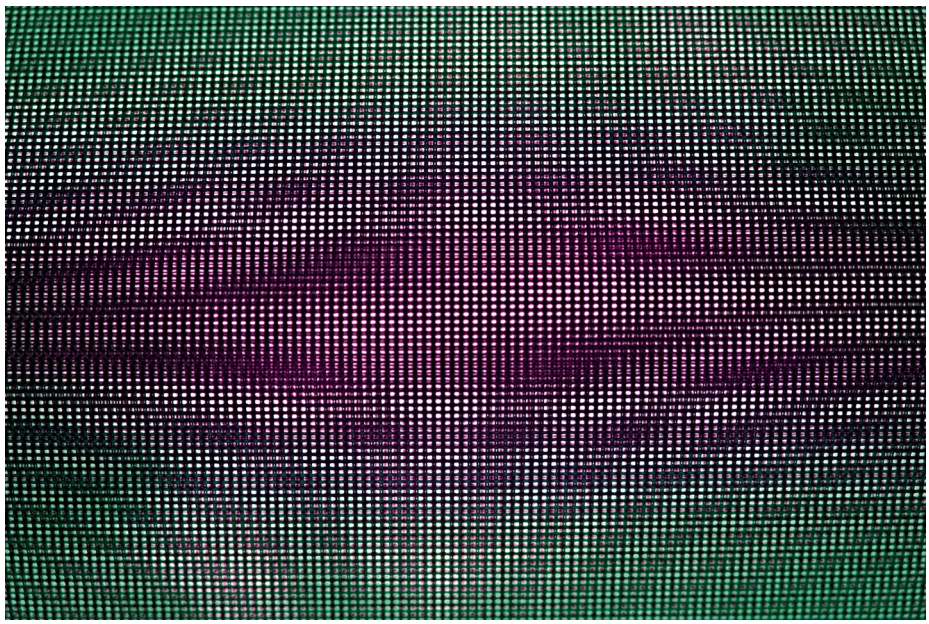
Computationally probing moiré magnets

We highlight two primary research papers, published in this issue of *Nature Computational Science*, on computational methods for moiré magnets.



When a two-dimensional (2D) lattice is stacked on top of another 2D layer with a small misorientation, it can create incommensurate moiré patterns, namely a moiré lattice. The twisted moiré structures can generate unprecedented electron correlations that can lead to exotic materials phenomena, such as the appearance of superconductivity[1]. Recently, the exploration of moiré bilayers has been expanded from graphene to more complex 2D materials, such as the magnetic material CrI_3 (ref. 2), which has given rise to many interesting phenomena that require advanced theories and computational methods for understanding the underlying mechanism. More importantly, given the existence of a large number of 2D candidates, robust computational predictions can also improve the design efficiency in such a vast space of materials.

The application of conventional electronic methods — such as density functional theory (DFT) — to study moiré physics poses many obstacles that are in part rooted in the superperiodicity[3] of moiré patterns resulting from twisting. Quantum mechanical calculations are usually performed with the assumption that certain periodicity exists in materials, and thus, a small supercell with periodic boundary conditions can be used to simulate the bulk material. In a twisted moiré lattice, however, the superperiodicity requires a larger supercell size for the simulation, which substantially increases the computational cost. For instance, as discussed in a News & Views by David Soriano, the moiré supercell can contain 50,000 atoms for twisted structures with small angles.

The magnetic interactions in moiré lattices add yet another complexity when studying these systems. Such interactions can bring about appealing physical behaviors of technical interest, such as skyrmions, which are noncollinear spin textures that could provide the next generation of information storage with low energy consumption. However, the addition of magnetic interactions in the total Hamiltonian can substantially increase the computational cost since the spin degree of freedom for accounting for magnetism in electronic structure methods requires additional self-consistent iterations to be solved. Therefore, effectively modeling large-scale magnetic systems for electronic structure methods remains daunting.

Notably, modeling exchange interactions in moiré magnets is a computationally expensive task, since the large commensurate moiré supercell can introduce many non-equivalent magnetic interactions. For instance, to model the 50,000 atoms for a moiré supercell, the resulting number of exchange parameters can go up to approximately 200,000, which is nearly impossible for conventional electronic structure methods to compute. To address this issue, a Brief Communication from Baishun Yang and colleagues introduced a microscopic moiré spin Hamiltonian to describe non-equivalent moiré magnetic exchange iterations (MMEIs) via a sliding-mapping approach. Instead of directly calculating the vast number of MMEI parameters, the authors mapped the MMEIs in twisted bilayer 2D magnets onto the interactions of well-known local stacking configurations — created by a smooth sliding process — in untwisted structures, thus making the calculation of such configurations affordable for DFT calculations. With the proposed method, the authors computationally uncovered that MMEIs can give rise to exotic properties, such as magnetic skyrmion bubbles, inspiring further experimental validations. It is worth noting that the sliding-mapping approach is applicable for moiré structures with small angles corresponding to large moiré lengths, which usually require larger supercells — and thus more computational resources — to be computed.

The computational challenges in studying moiré magnets can also be addressed with the help of machine learning. A deep learning-based approach for representing the DFT Hamiltonian was shown to reduce the computational cost for large-scale materials simulations, but its application was mostly limited to non-magnetic systems[4]. In a Brief Communication published in this issue, He Li and colleagues developed an extended deep-learning DFT Hamiltonian (xDeepH) method to learn the spin–orbital DFT Hamiltonian on atomic and magnetic structures. To reduce the overall training cost, they incorporated prior physical knowledge to the neural network model, including the principles of locality in electronic structure and symmetry operations. xDeepH was applied to several examples in which the same calculation using conventional

DFT is overly expensive. For CrI_3, the authors theoretically demonstrated that their model can be transferrable across twisted magnetic superstructures, even capturing magnetic skyrmions based on the twist angles.

As highlighted by Soriano, computations at the DFT-accuracy level for complex magnetic systems will allow scientists to investigate magnetic superlattices more systematically, which will ultimately help to improve the understanding of the underlying physics and to make reliable predictions of new moiré magnets with exotic properties. Exciting times ahead!

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