Using twist angle selection and the transition structure factor to speed up finite size convergence in periodic coupled cluster theory

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There is a current need in material design for benchmarks from high accuracy methods. One such method, coupled cluster theory, show potential for use in benchmarking solids, but suffers from large errors known as finite size effects. These finite size effects come from modeling a solid with a finite size supercell and causes a slow, polynomial scaling to the thermodynamic limit (TDL). Here, I will present on our new twist angle selection method called "structure factor twist averaging" that was developed to help reduce these finite size effects and speed up convergence to the TDL [J. Chem. Phys. 150, 191101 (2019), Nat Comput Sci 1, 801–808 (2021)]. Twist angles are offsets to the k-point grid that break the degeneracy of the orbitals, results in a change to the occupation and energy of the system for each given twist angle. Averaging over a set of these twist angles, then, will result in a more balanced depiction of the overall system with less finite size effects, but comes with a significant scaling in cost. Our method reduces this cost of obtaining the average by finding the one twist angle that reproduces the average system and, therefore, has the smallest amount of finite size effects present. Our special twist angle is chosen by using a property of the correlation energy known as the transition structure factor. As this property is directly linked to the correlation energy of the system, I will show that this is an effective way to choose our special twist angle. In addition, I will show how we can use this new structure factor twist averaging scheme to calculate properties of solids, such as energy differences between phases of metals and the semiconductor-to-metal transition pressure for two phases of silicon [Nat Comput Sci 1, 801–808 (2021)].