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QUANTUM CHEMISTRY

Deep learning chemistry ab initio

> Our approach finds very accurate neural network solutions for the much harder case of electrons — fermions that have to obey the Pauli exclusion principle

Understanding how electrons distribute themselves around nuclei is probably one of the biggest challenges in chemistry. Schrödinger gave us one of the most elegant mathematical formulations of this problem, the solution of which gives access to the deepest secrets of atomic and molecular behaviour. Although it is possible to solve this equation analytically for a single-electron system, things become complicated when the number of electrons increases. The race for methods to get closer to the exact solution to the Schrödinger equation for multiple electrons has been non-stop since its formulation. Jan Hermann, Zeno Schätzle and Frank Noé report in *Nature Chemistry* a new approach, PauliNet, which uses deep learning quantum Monte Carlo to find multidimensional wavefunctions that describe molecules with up to 30 electrons.

Advances in computational chemistry are often motivated by the search for methods that are either more accurate or computationally efficient. Although approaches such as configuration interaction, coupled cluster and quantum Monte Carlo afford highly accurate values of the electronic energy, their computational costs increase exponentially with the number of electrons. Approximations of the Schrödinger equation are therefore necessary if we want to look at molecules beyond hydrogen. Methods such as density functional theory require much less computer power than the methods mentioned above at the price of a much lower accuracy.

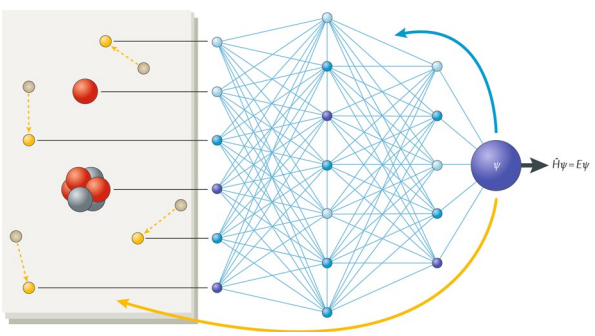
Machine learning has offered another way to explore chemical space. A way that is much more empirical: the algorithm can predict properties or new materials based on knowledge gained about known systems, without the need for solving the Schrödinger equation. By contrast, Noé and colleagues used a deep neutral network to directly represent the electronic wavefunction and provide an ab initio solution of the Schrödinger equation. "Giuseppe Carleo and Matthias Troyer showed that such an approach is possible by solving the bosonic Schrödinger equation for lattice systems using neural networks," says Noé. "Our approach finds very accurate neural network solutions for the much harder case of electrons — fermions that have to obey the Pauli exclusion principle. This is the very reason that solving the electronic Schrödinger equation is hard. That is why we call our neural network PauliNet."

Chemists use Slater determinants to approximate a multi-electron wavefunction in a way that obeys the Pauli principle. However, because we do not know the exact wavefunction, we need a very high number of Slater determinants to get a close approximation, which makes accurate quantum calculations extremely computationally demanding. The team used a 'backflow' approach to optimize the number of Slater determinants necessary, starting with an approximate Slater determinant and calculating a baseline wavefunction. The information from the baseline wavefunction is fed into a new Slater determinant to obtain a new, more accurate wavefunction and the process is iterated to obtain a desired accuracy. "The backflow idea has been used in quantum chemistry but backflow functions were not flexible enough to be incorporated into machine learning. In PauliNet, we developed a new type of neural network backflow function that is easy, far more powerful than established backflows, which get accurate solutions with a handful of Slater determinants where we otherwise would have required thousands," explains Hermann.

The team tested PauliNet on Be and B atoms, H_2 and LiH molecules, the linear chain H_{10}, which is characterized by a strong electronic correlation that usually requires a large number of Slater determinants to be properly calculated, and on the energy barrier of the automerization of cyclobutadiene. For all these systems, PauliNet could reach between 97 and 99.9% of the correct energy using only between 6 and 16 Slater determinants.

One big advantage of PauliNet is that its computational cost does not grow as much as other more common, high-accuracy approaches with the number of electrons. Methods such as configuration interaction or coupled cluster can scale with N^6–N^7, depending on the variant, whereas PauliNet is expected to scale with N^4. "We believe the sweet spot of our method is small molecules or material unit cells with 50–200 electrons. There is currently no black-box quantum chemistry method that can solve such systems to arbitrary accuracy," says Noé. Accurate quantum chemistry descriptions of small molecules at a reasonable computational cost will serve as solid training sets for 'more empirical' machine-learning approaches to explore the vastness of chemical space.

Gabriella Graziano



Credit: Adapted from Hermann, J., Schätzle, Z. & Noé, F. *Nat. Chem.* https://doi.org/10.1038/s41557-020-0544-y (2020), Springer Nature Limited

ORIGINAL ARTICLE Hermann, J., Schätzle, Z. & Noé, F. Deep-neural-network solution of the electronic Schrödinger equation. *Nat. Chem.* https://doi.org/10.1038/s41557-020-0544-y (2020)
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