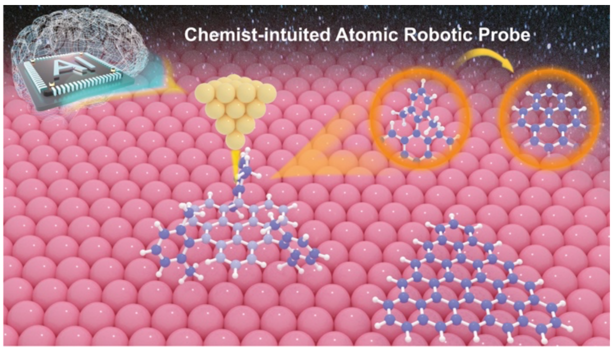
TITLE: AI for carbon-based quantum materials

Jie Su[1], Jiali Li[1], Xiaonan Wang[2], Jiong Lu[1]
1. National University of Singapore, Singapore, Singapore.
2. Department of Chemical Engineering, Tsinghua University, Beijing, China.

Atomic-scale manufacturing of carbon-based quantum materials with single-bond precision holds immense potential in advancing tailor-made quantum materials with unconventional properties, which are crucial in developing next-generation spintronic devices and quantum information technologies. To date, on-surface chemistry approaches, including surface-assisted synthesis and probe-assisted manipulation, while demonstrating huge potential in fabricating these materials, are impeded by the challenges of reaction selectivity control, or restricted by scalability and production efficiency, severely hindering their practical deployment. Herein, we demonstrate the concept of the chemist-intuited atomic robotic probe (CARP) by integrating probe chemistry and artificial intelligence, allowing for conducting precise molecular surgery, aiming for the on-demand synthesis and comprehension of the elusive carbon-based quantum matters with single-bond precision. Such a concept is validated by a novel multi-bond molecular surgery to fabricate open-shell magnetic nanographenes, known as single-molecule quantum π-magnets (SMQMs), with tunable topology and spin multiplicity triggered by site-selective probe chemistry. Our deep neural networks not only transform such a complex probe-chemistry issue into machine-understandable tasks but also provide valuable chemist-intuition into elusive reaction mechanisms by extracting the critical chemical information within the data. A joint experimental and theoretical investigation demonstrates that a voltage-controlled two-electron-assisted electronic excitation enables synchronous six-bond transformations to extend the zigzag topology of SMQMs, triggered from the phenyl *sp2* -CH bond activation, aligning with initial conjectures given by the deep neural models. Our work represents a significant leap from autonomous fabrication to intelligent synthesis with unparalleled levels of selectivity and precision beyond the current synthetic tools, thus creating a major paradigm shift in the on-demand manufacturing of novel organic quantum materials for on-chip integration.



The innovative CARP framework enables the microscope to "think" and "act" as an experienced surface chemist at the atomic scale

Reference

1. Nature Synthesis 3, 466–476 (2024)
2. J. Am. Chem. Soc. 143 (27), 10177-10188 2021