

Text driven ab initio simulations of chemical reactions using large language models

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Ab initio atomistic simulations are important for understanding reaction energetics, mechanisms, and kinetics, but practical workflows often require substantial manual effort in model preparation, input file generation, and post-analysis. In this work, we present a text driven program that combines large language model (LLM) assisted task planning with local scripts to automate ab initio atomistic simulation workflows from natural language prompts. The program decomposes user requests into chemically meaningful calculation tasks, prepares input parameters for ab initio calculations, runs calculations on an HPC, and performs post-analysis to generate a report. The workflow emphasizes both reliability and data security. Instead of asking an LLM to directly analyze confidential data, such data are randomized locally before being uploaded. The LLM is then used to generate analysis scripts, which are applied only to the true data on local machines. This strategy reduces hallucination risk and protects sensitive simulation results. In addition, a user-friendly web interface has been established to make the program accessible for task submission, workflow monitoring, and report generation.

To validate the developed program, two practical examples from the literature are demonstrated. First, reaction mechanisms of Al₂O₃ atomic layer deposition are reproduced using only a simple text prompt, such as: “Calculate the reaction pathway of Al(CH₃)₃ + Al(OH)₃ → Al(OH)₂OAl(CH₃)₂ + CH₄ using the B3LYP D3 def2-TZVPP method.” Second, reaction-rate constant calculations are demonstrated for SiH₂Cl₂ → SiCl₂ + H₂ between 300 and 1000 K using mixed levels of theory: B3LYP D3 cc-pVTZ for geometry optimization and transition-state search, combined with DLPNO-CCSD(T) cc-pVQZ single-point energies. These examples show that time-consuming and complex ab initio simulation workflows can be automated using intuitive text inputs.