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LLM Agents for Autonomous Density Functional Theory Simulations from Natural Language

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Motivation

Challenges in High-Throughput DFT

- Automation is fragile: Traditional workflows easily fail when unexpected issues occur.
- Expertise barrier remains: Users still need deep technical knowledge to set up and troubleshoot simulation.
- Inefficient resource use: Static pipelines waste resources by not adapting based on intermediate results.

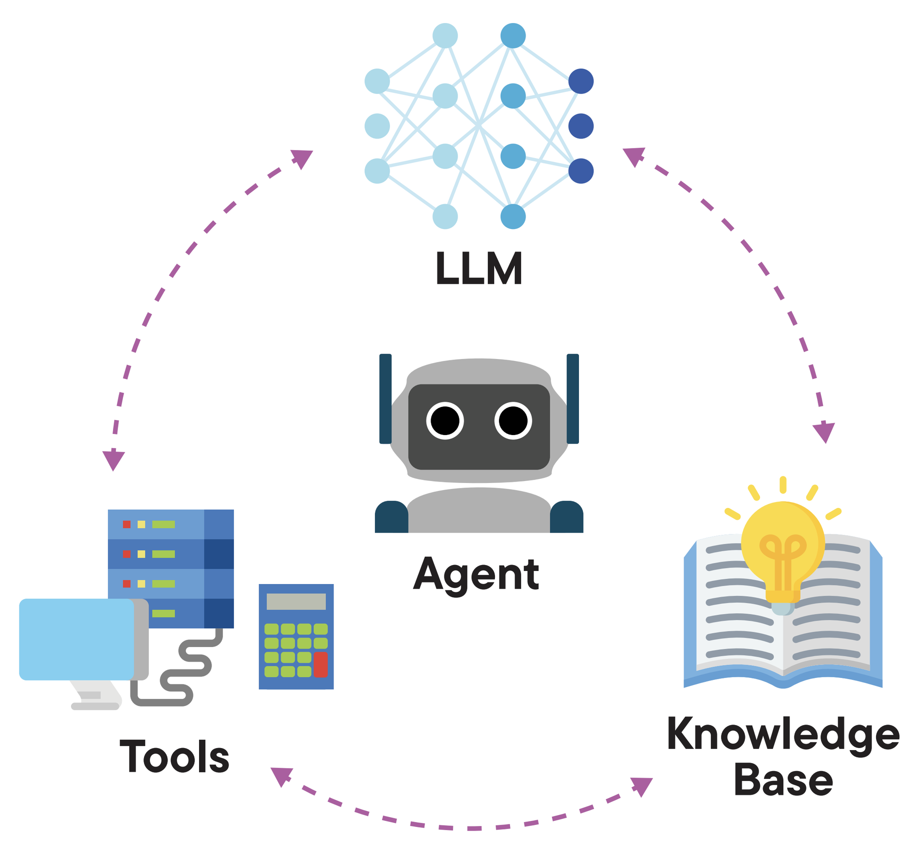
Introduction - From Automated to Autonomous

LLM Agents

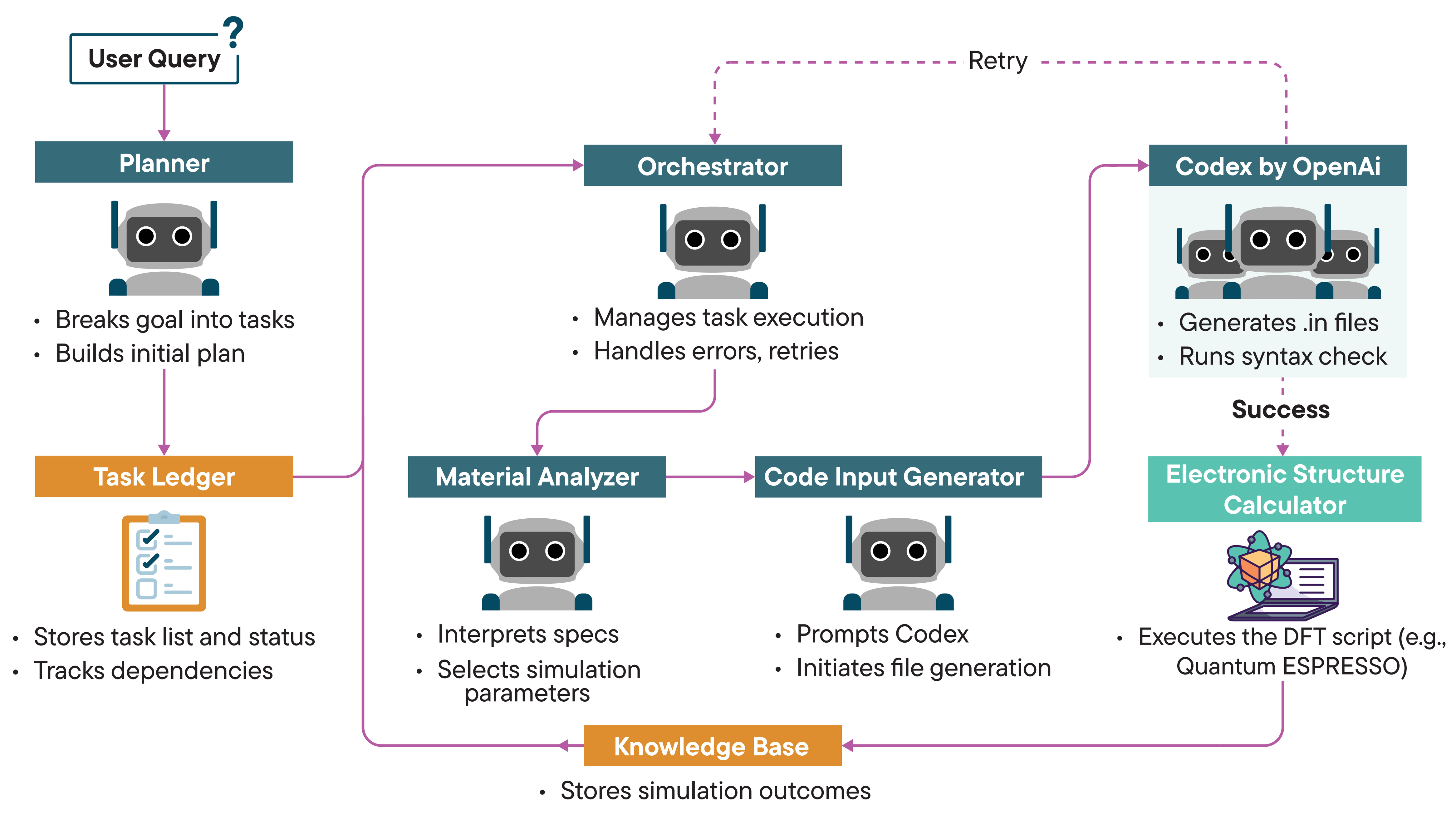
- LLM agents are customized for specific subtasks within a broader scientific goal, each guided by a focused system message and tailored instructions.
- This specialization enables accurate execution - such as planning, input generation, error correction, and analysis - and supports modular, dynamic workflows.

Agency and Autonomy

- Agency allows LLMs to reason over time, make decisions, and adapt plans based on intermediate results rather than following a fixed sequence.
- Dynamic workflows built from agents recover from failures, reprioritize tasks, and optimize computational resources in real time.
- Transitioning from automated to autonomous systems greatly expands the scalability and efficiency of scientific discovery.



Agentic Setup



LLM Agents — Ledgers
Third-Party Agent — Tools

Key Agenic Benifits

- Enables complex DFT workflows to be executed autonomously, without brittle hard-coded pipelines.
- Supports real-time error recovery, task reprioritization, and reflection - capabilities beyond traditional automation.
- Modular design allows integration of agents built by other researchers, labs, or companies for specialized tasks.
- Demonstrates how LLM agents can collaborate to solve scientific problems adaptively, scalably, and without retraining.
- Can be encapsulated as a single agent module within a larger catalyst discovery framework.

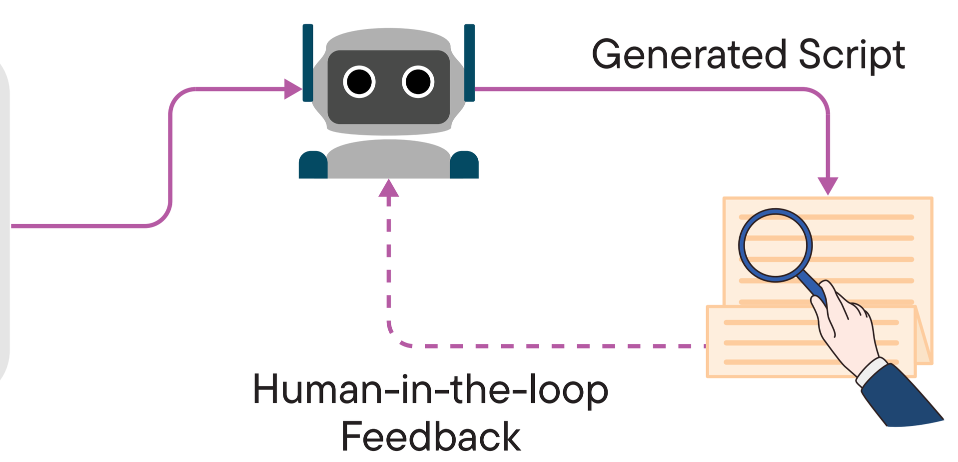
Case Study - Carbon Dioxide Reduction

CO_2 Adsorption Energy on a Cu(111) Slab

- CO_2 adsorption is the first step in electrochemical CO_2 reduction (CO_2RR).
- Adsorption energy affects both activity and selectivity of the catalyst.
- Simulating CO_2 on Cu(111) provides insight into surface–molecule interactions.

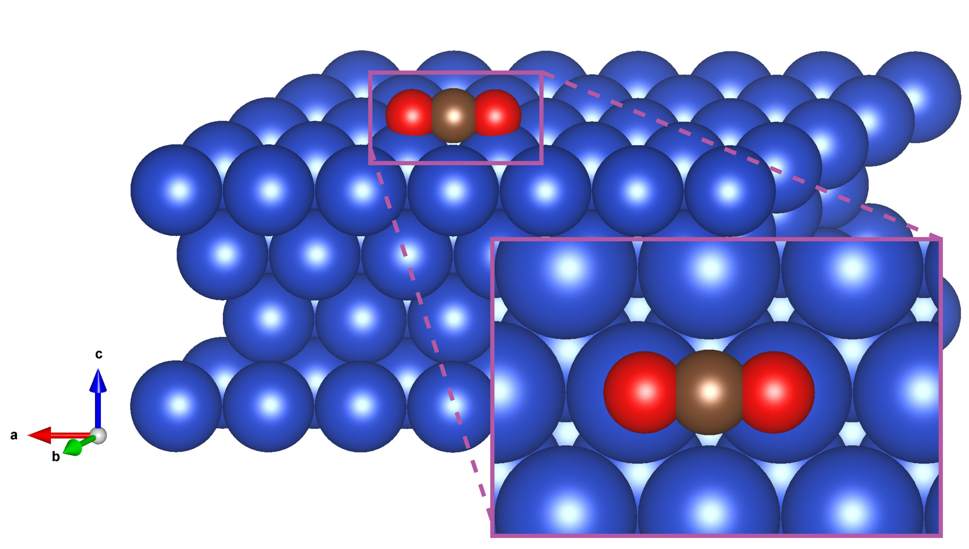
User Query

> Make me a Quantum ESPRESSO simulation file to relax a CO_2 molecule adsorbed at the bridge site of a Cu(111) slab using the rPBE exchange-correlation functional...



Output

```
&control
  calculation='relax',
  prefix='Cu111_CO2',
  pseudo_dir = '/app/pseudos'
/
&system
  ibrav=0,
  nat=99,
  ecutwfc=30.0,
  ecutrho=240.0,
  occupations='smearing',
  smearing='gaussian',
  degauss=0.002,
```



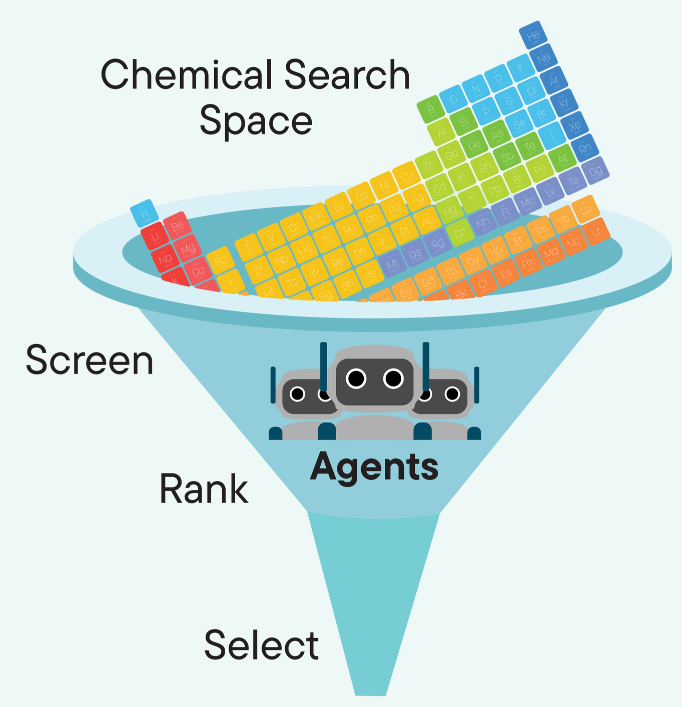
Generated Geometry: CO_2 on Cu(111) - bridge site

Simulation based on: Wang et al. Energy Environ. Sci., 2023,16, 4388-4403

Conclusions

- LLM agents can autonomously interpret scientific queries and generate DFT input files.
- Agentic workflows enable autonomous input generation, real-time error recovery, and adaptive task management—surpassing traditional scripted automation.
- Modular agent design supports reuse, scaling, and integration into broader catalyst discovery platforms.

Next Steps



- Embed this agent within a broader LLM system to selectively run only essential DFT simulations during high-throughput catalyst screening.
- Use LLM reasoning to decide when DFT is actually needed, minimizing total simulations while exploring materials space more intelligently.