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Integrating Autonomous Systems for Advanced Material Discovery: Bridging Experiments and Theory Through Optimized Rewards

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Materials Science cannot change overnight….

… but what is happening now comes very close

The inflection point (for theory): 2006

Predicting crystal structure by merging data mining with quantum mechanics

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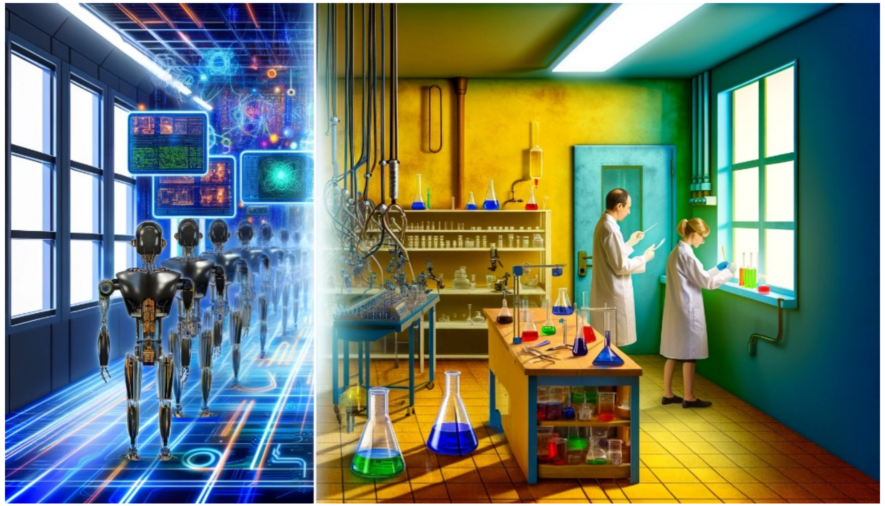
Publication by Gerd Ceder paper that is broadly seen as the inflection point launching Materials Genome Initiative in US and equivalent programs worldwide

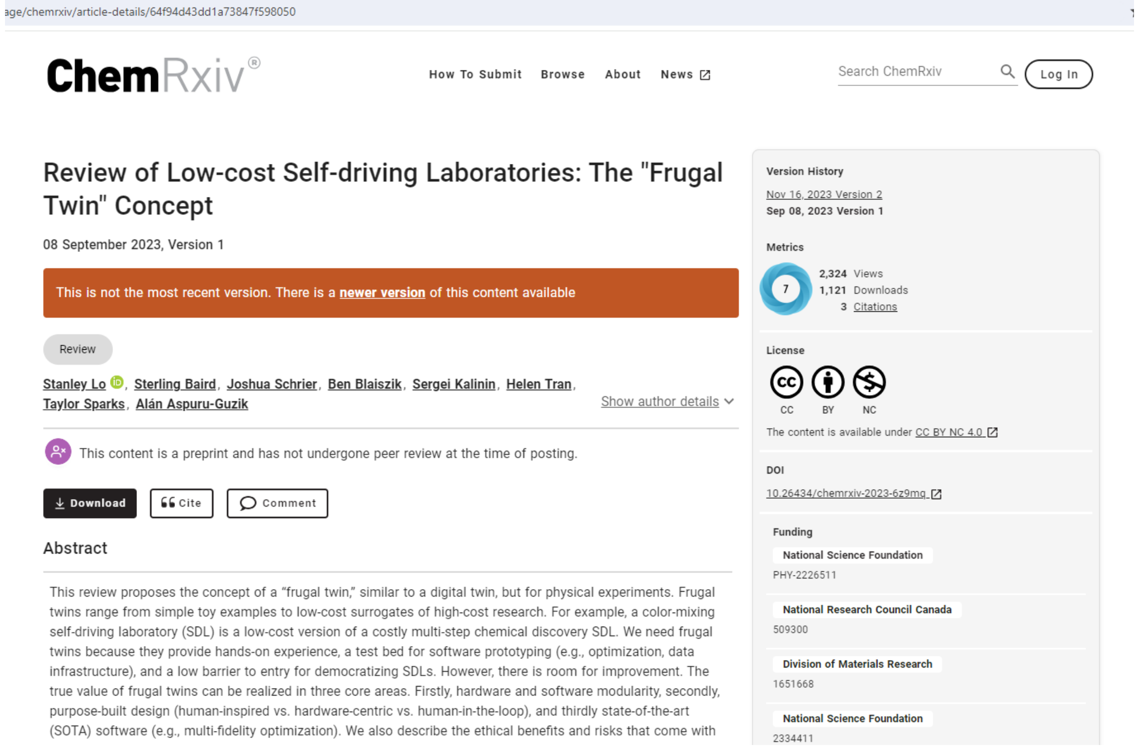
Launch of AWS (Amazon Web Services) made cloud computing a reality – allowing businesses and scientists alike have access to computational resources without the need to build and maintain clusters



Machine learning in theory:
- Homogeneous workflows
- Known causal structure/lack of exogeneous factors
- Requires know-how, but relatively low entry barrier
- Easy to scale (given the funding)

The inflection point (for experiment): ~2020

- **Before 2010:** A number of (usually) confidential efforts in industry
- **2010 - 2015:** Early adopters and visionaries (Cronin, Maryama, Kusne, etc)
- **2015 – 2020:** The time of engineers
- **2020 – now:** Automated experimentation becomes broadly available with very low cost entry barriers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Review of Low-cost Self-driving Laboratories: The "Frugal Twin" Concept

08 September 2023, Version 1

This is not the most recent version. There is a newer version of this content available

Review

Stanley Lo, Sterling Baird, Joshua Schrier, Ben Blaiszik, Sergei Kalinin, Helen Tran, Taylor Sparks, Alán Aspuru-Guzik

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This content is a preprint and has not undergone peer review at the time of posting.

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Abstract

This review proposes the concept of a "frugal twin," similar to a digital twin, but for physical experiments. Frugal twins range from simple toy examples to low-cost surrogates of high-cost research. For example, a color-mixing self-driving laboratory (SDL) is a low-cost version of a costly multi-step chemical discovery SDL. We need frugal twins because they provide hands-on experience, a test bed for software prototyping (e.g., optimization, data infrastructure), and a low barrier to entry for democratizing SDLs. However, there is room for improvement. The true value of frugal twins can be realized in three core areas. Firstly, hardware and software modularity, secondly, purpose-built design (human-inspired vs. hardware-centric vs. human-in-the-loop), and thirdly state-of-the-art (SOTA) software (e.g., multi-fidelity optimization). We also describe the ethical benefits and risks that come with

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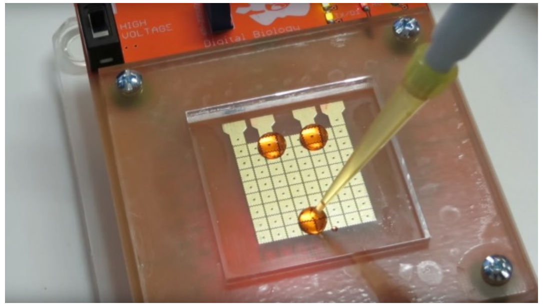
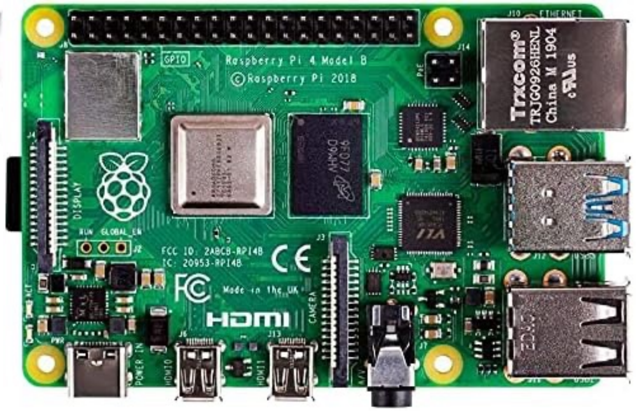
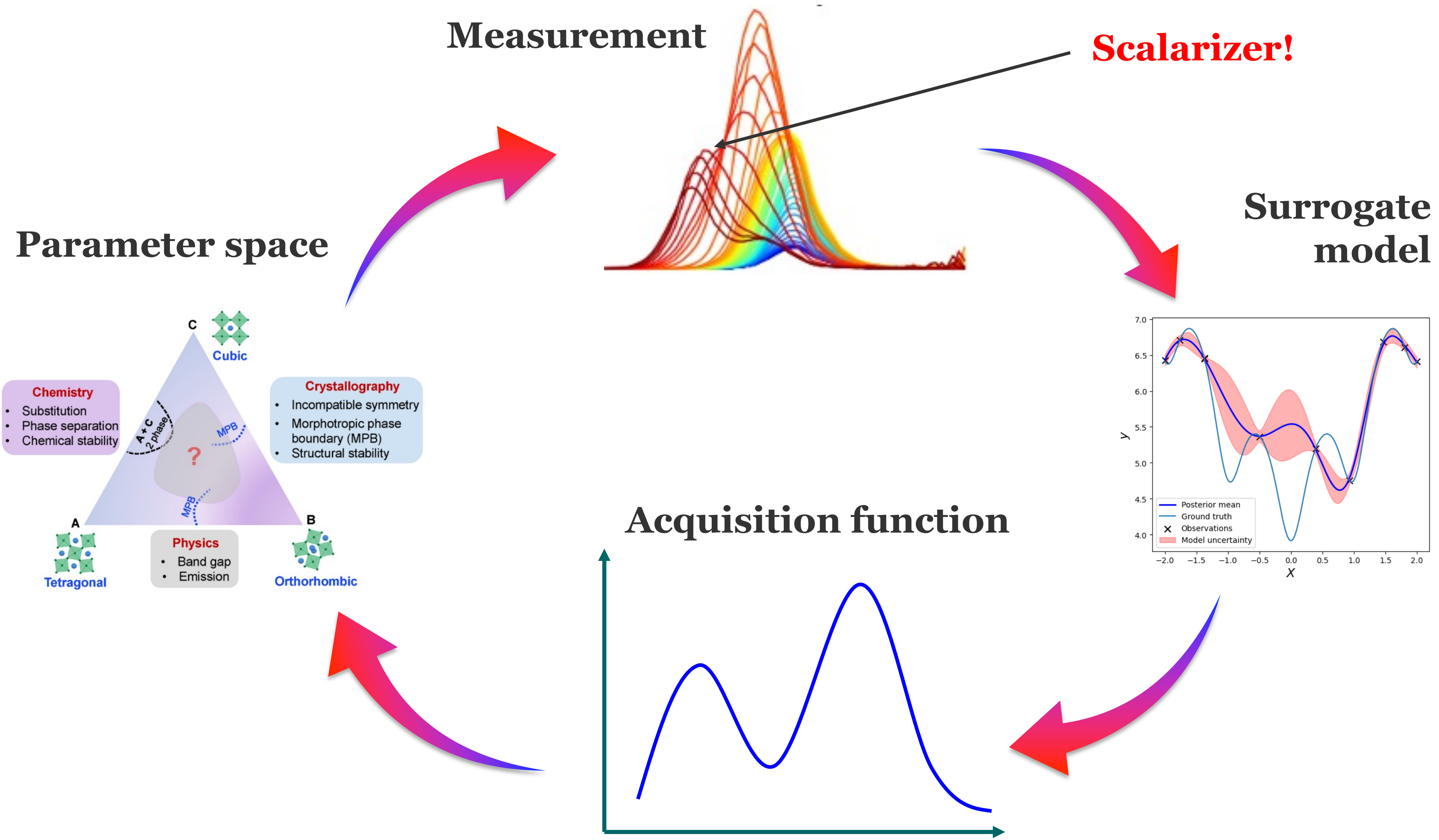


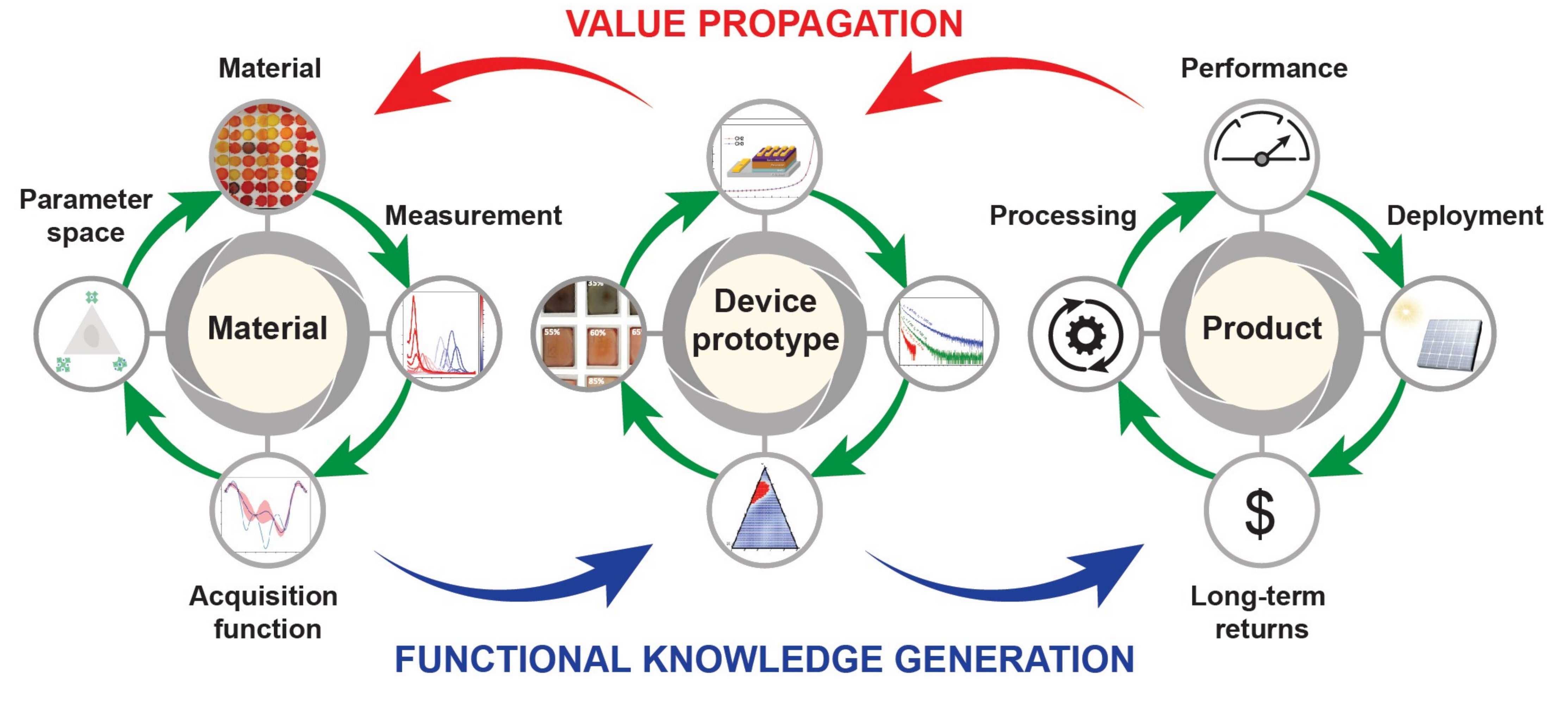


Classical Bayesian Optimization



Implicitly, we postulate that cost and gain are constant along the iterative cycle

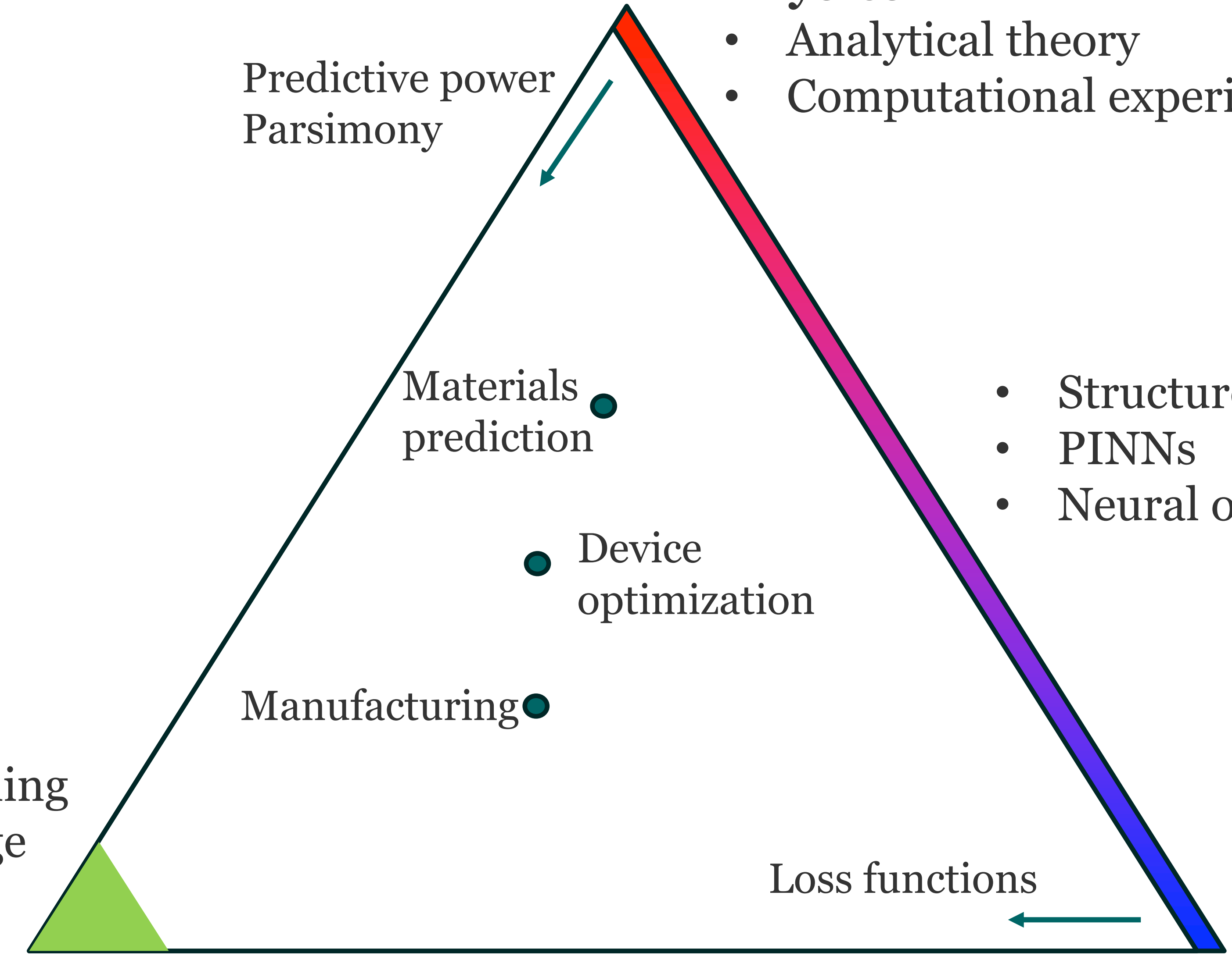
Multiple connected cycles



- Scientific data is often high-dimensional data sets
- The value of which can only be understood down the line
- R&D process involves multiple coupled exploration loops

Is Machine Learning and Physics Enough?

Physics
- Analytical theory
- Computational experiments

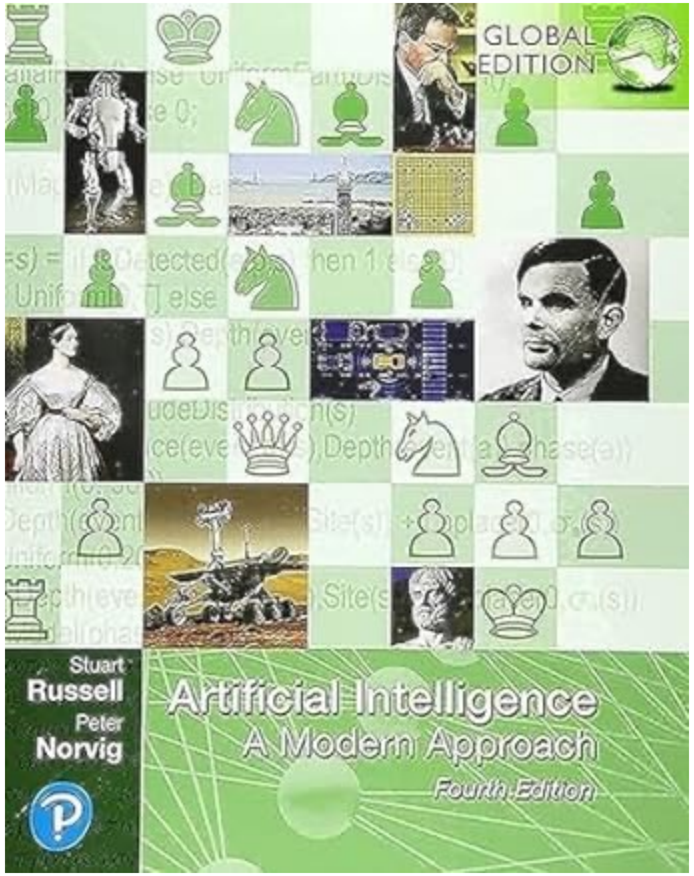


- Structured GPs
- PINNs
- Neural operators

Rewards
- Global warming
- Better storage materials
- …

Data:
- DCNNs
- LLMs
- And so on

Somewhat remarkably, almost all AI research until very recently has assumed that the performance measure can be exactly and correctly specified in the form of utility or reward function



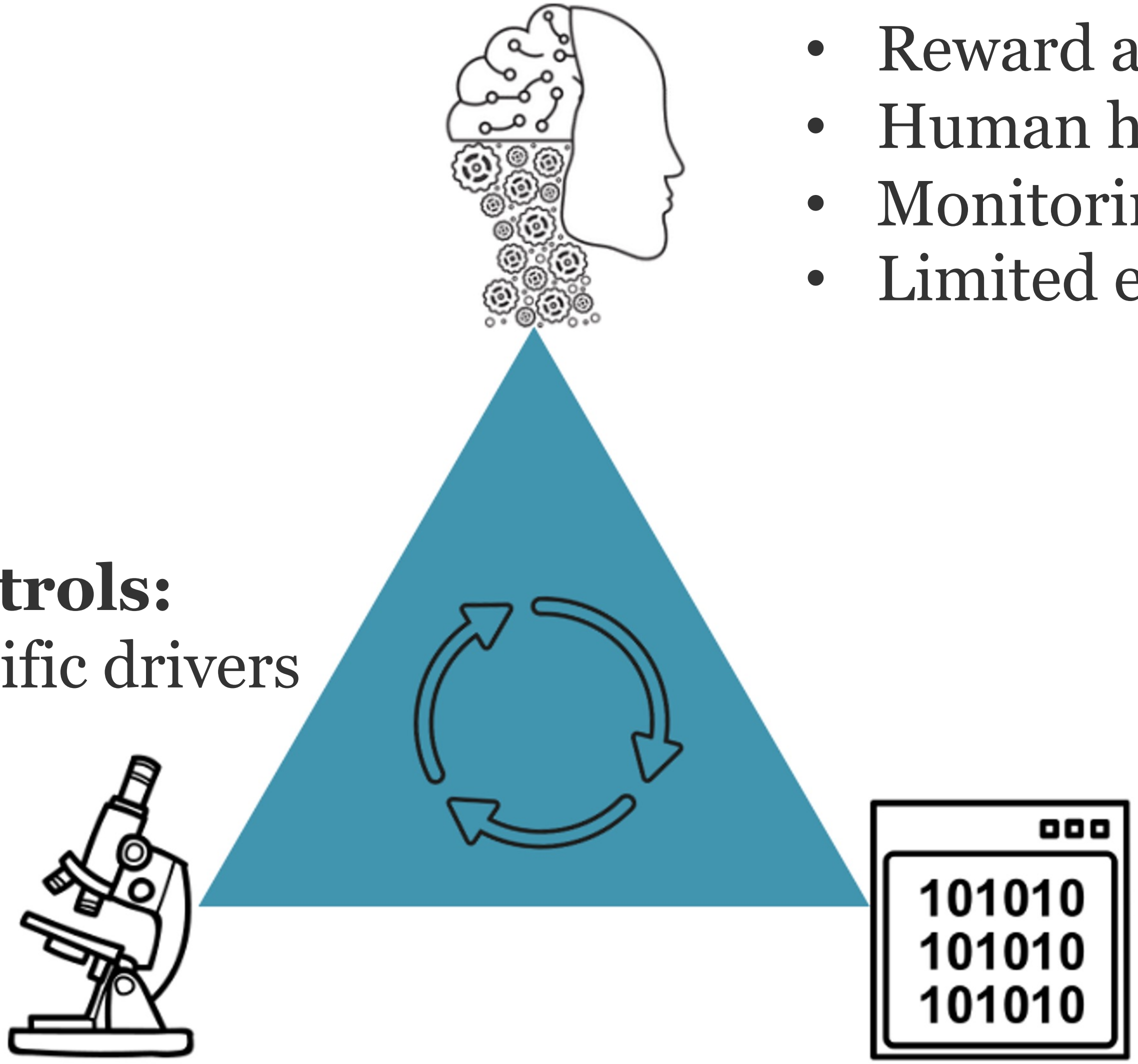
Challenges for the AE Microscopy

Workflow design:

- Reward and value functions
- Human heuristics
- Monitoring
- Limited experimental budget

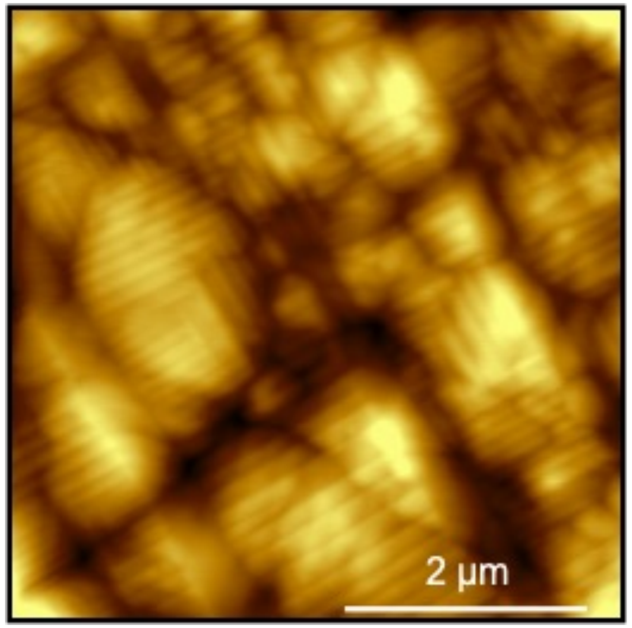
Engineering controls:

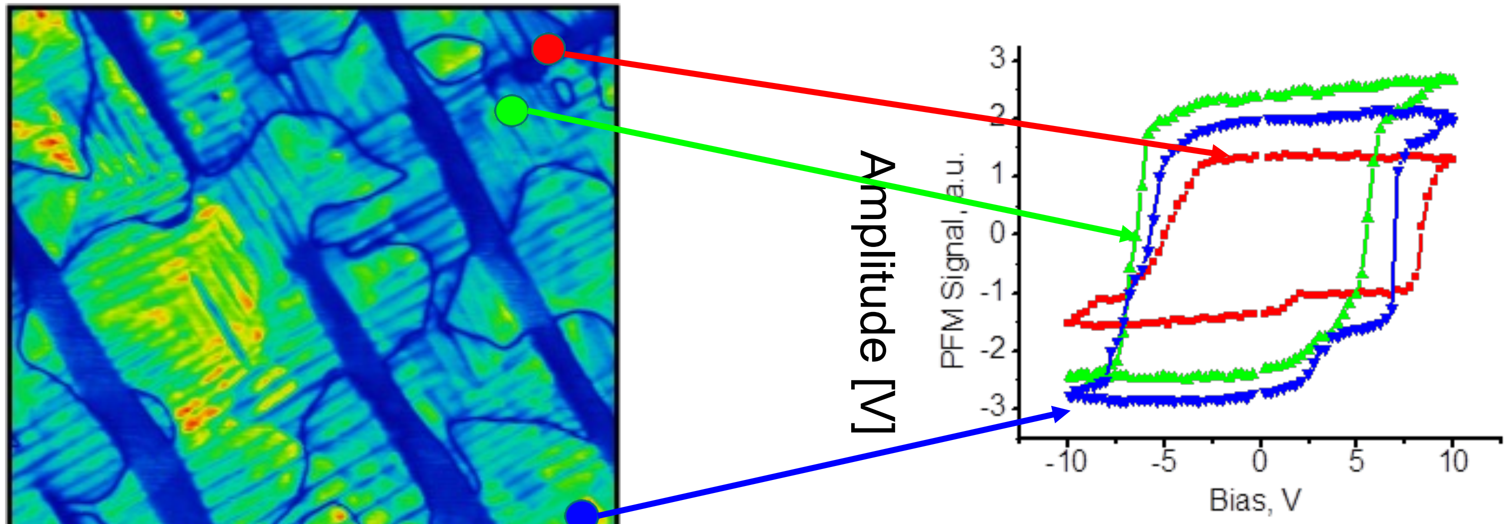
- Instrument-specific drivers
- Hyperlanguage
- Python APIs



ML/AI

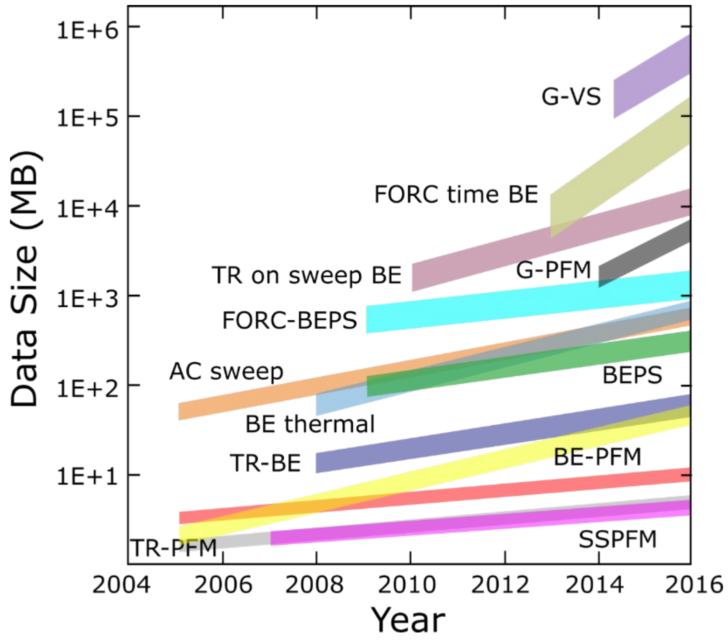
Why do we do experiments?

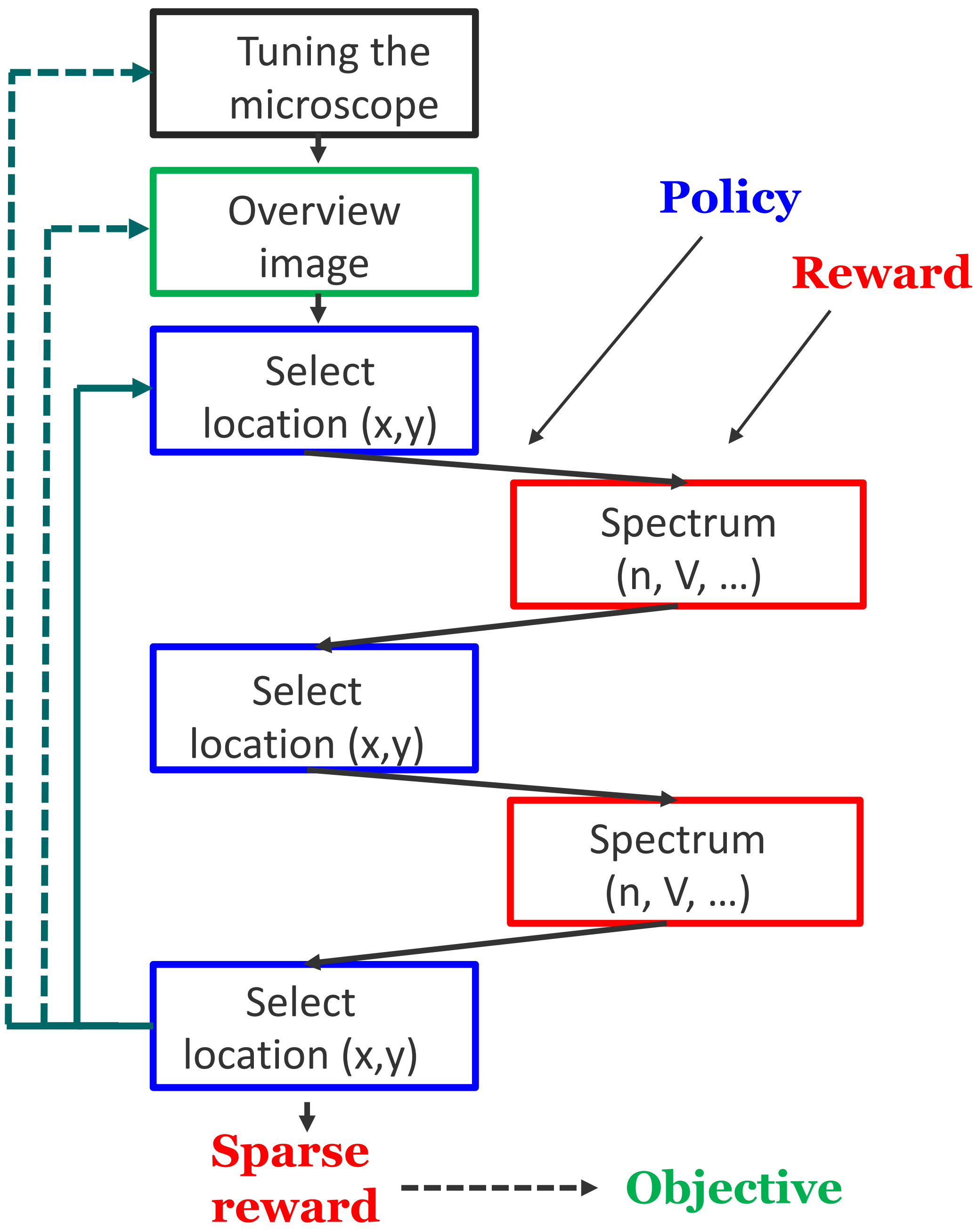




- Interesting functionalities are expected at the certain elements of domain structure
- We can guess some; we have to discover others

- **Experimental objectives → ML Rewards**
  - Microscope optimization
  - Properties of a priori known regions of interest
  - Discovery of regions with interesting properties
  - Physical theory falsification





To implement the ML workflows, we start from emulating the human operations:

- Well defined and explainable commands
- Extensive domain expertise
- Potentially available data from experiments

Development of ML workflows can give rise to more complex imaging modalities

- Data volumes and dimensionalities above human level
- More complex modes of sampling
- "Guardian angel" modules

However, we always have to think about

- Reward function(s) for imaging problem
- Reward functions for materials problem
- Overall objective

Reward functions in imaging

Imaging Optimization

Physical laws discovery

Image-based reward functions

- Human selected objects (DCNNs)
- Equal sampling of feature space
- Equal sampling of parameter space (combi library)

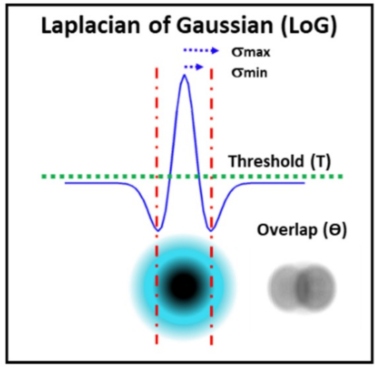
Structure property relationship discovery

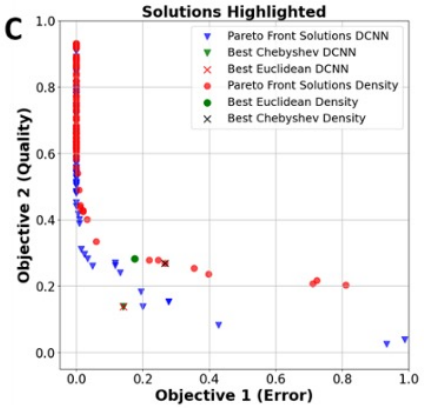
- Reward definition (with cost)
- Tuning curiosity
- Human in the loop DKL

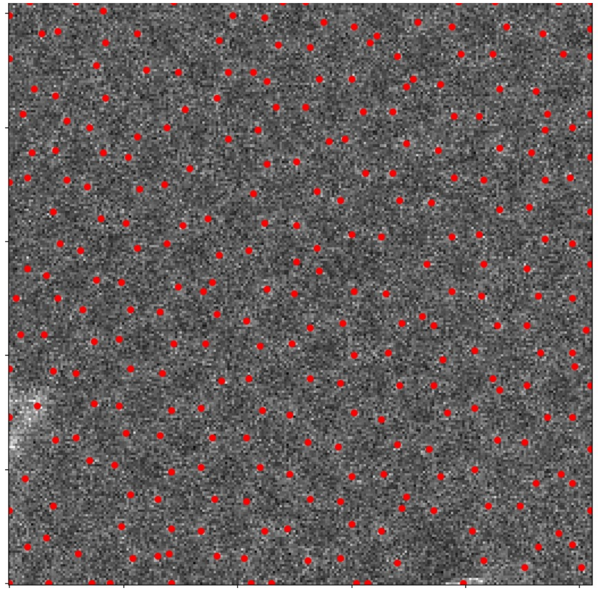
Co-orchestration multiple tools
Co-navigation between theory and experiment

Reward functions in imaging

Atom Finding







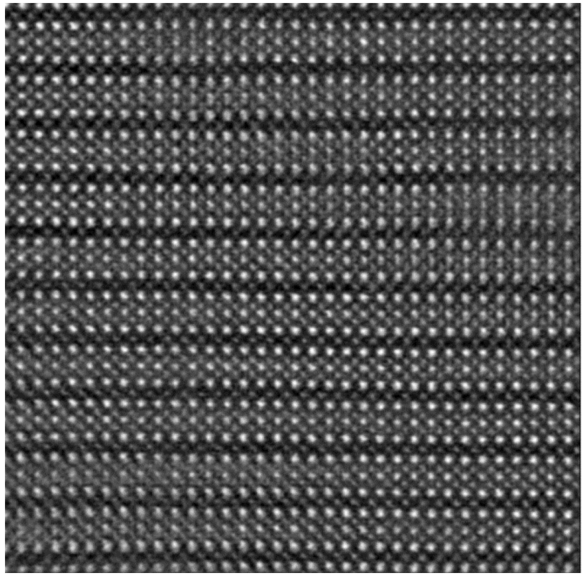
Rewards 1:
N atoms

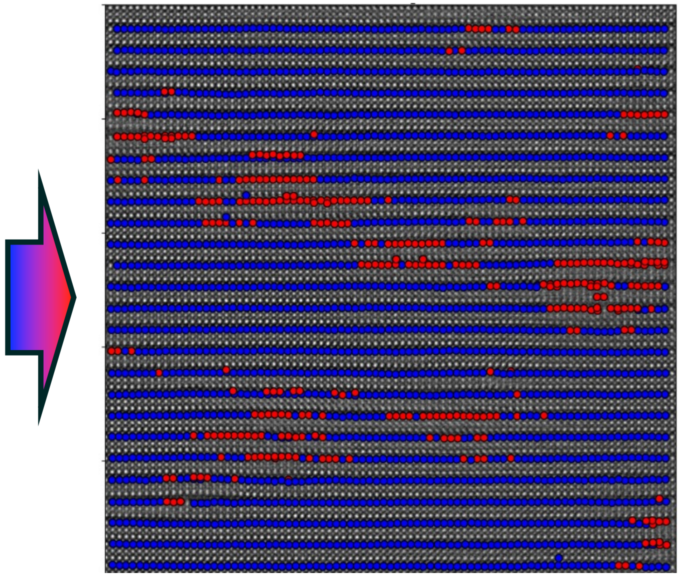
Reward 2:
Atomic
distances

Building Image Analysis Pipelines (non-myopic)

Image, video → Creating descriptors (size, shape) → [Identity | Radon | Fourier] → [GMM | PCA | NMF] → Visualization and descriptive statistics:

Loading maps, components

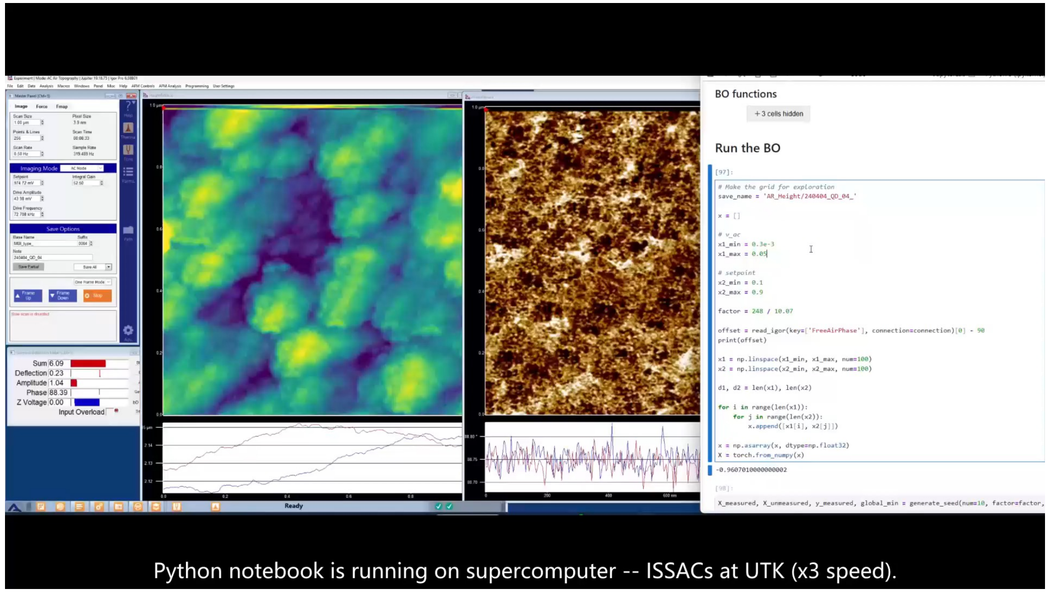




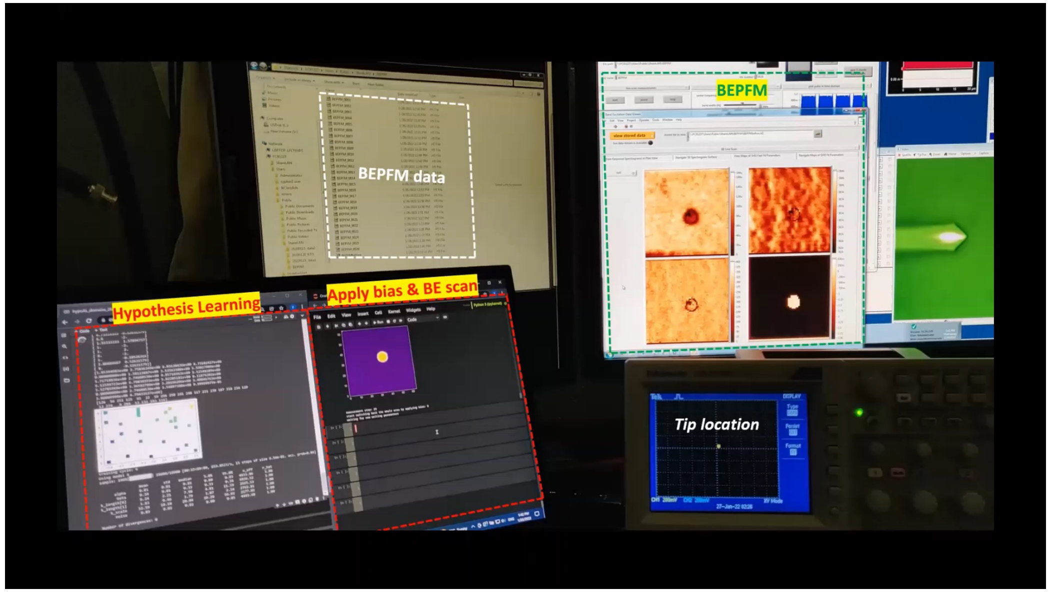
Reward function approach reduces the supervised learning problem to optimization problem (human labeling, sensitive to distribution shift, black box) -> (unsupervised, robust, explainable)

Reward functions in (actual) imaging

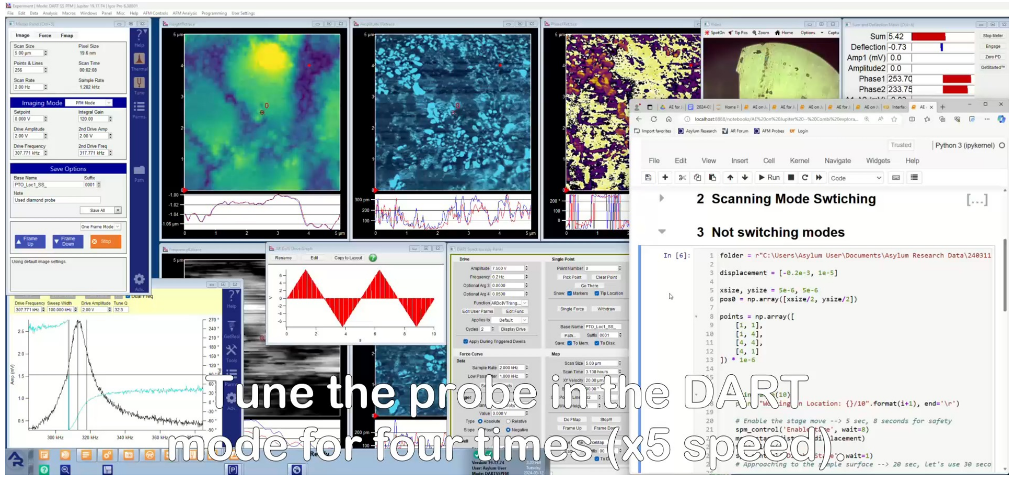
Optimization of imaging parameters



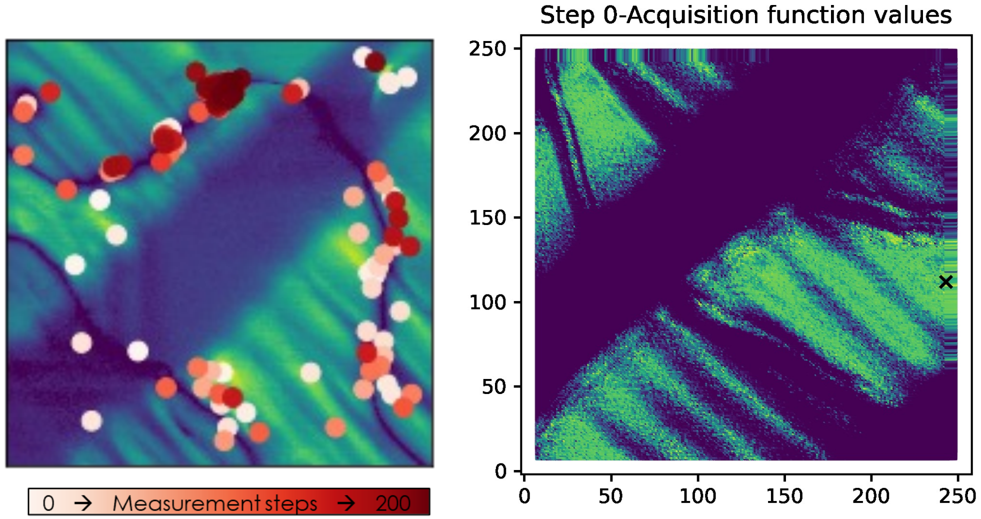
Discovery of physical laws



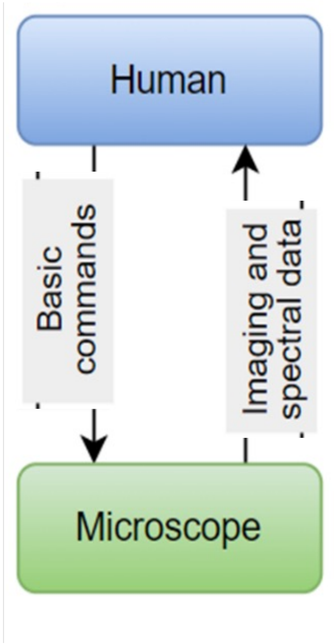
Exploring combinatorial libraries

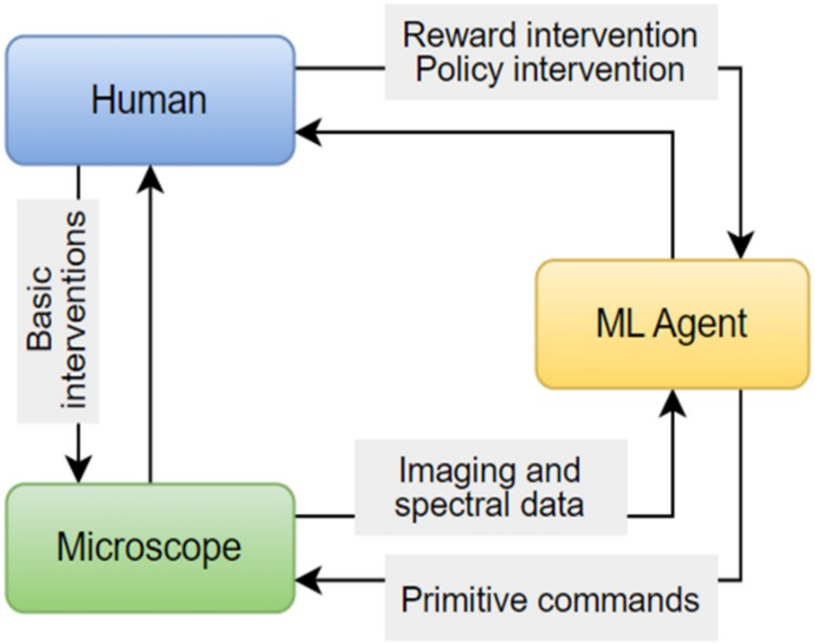


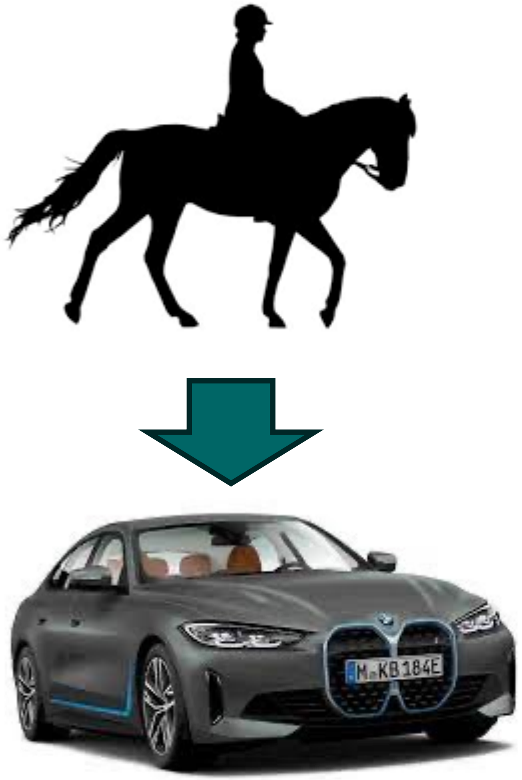
Discovering structure-property relationships



Human in the loop AE



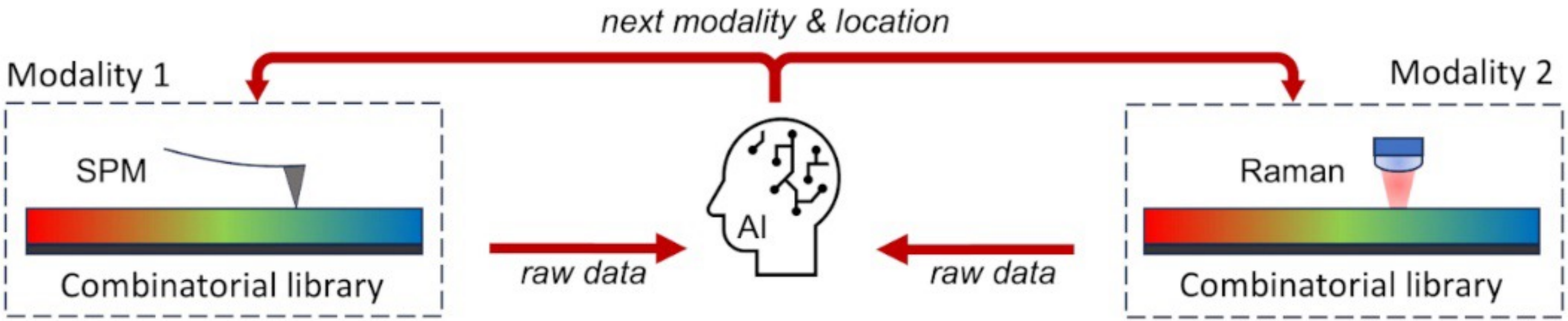


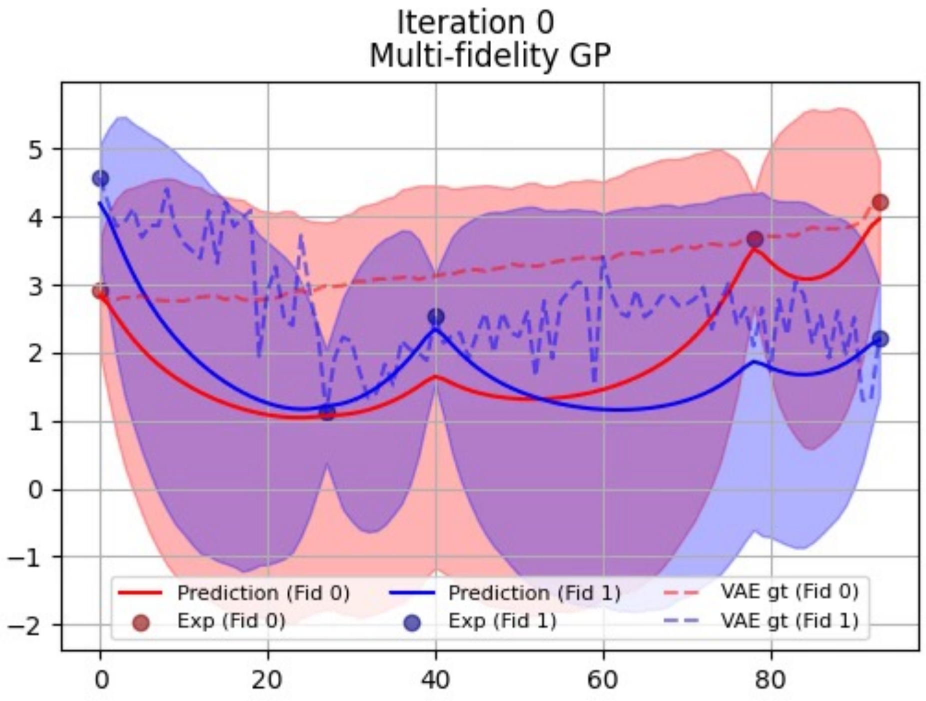


We can intervene on:

- Policies (acquisition functions): type and parameters
- Scalarizers (physics descriptors): type and parameters
- Knowledge injection
- Direct operation

Orchestrating multiple experiments





- Co-orchestration of the Raman and PFM experiments on the pre-acquired data

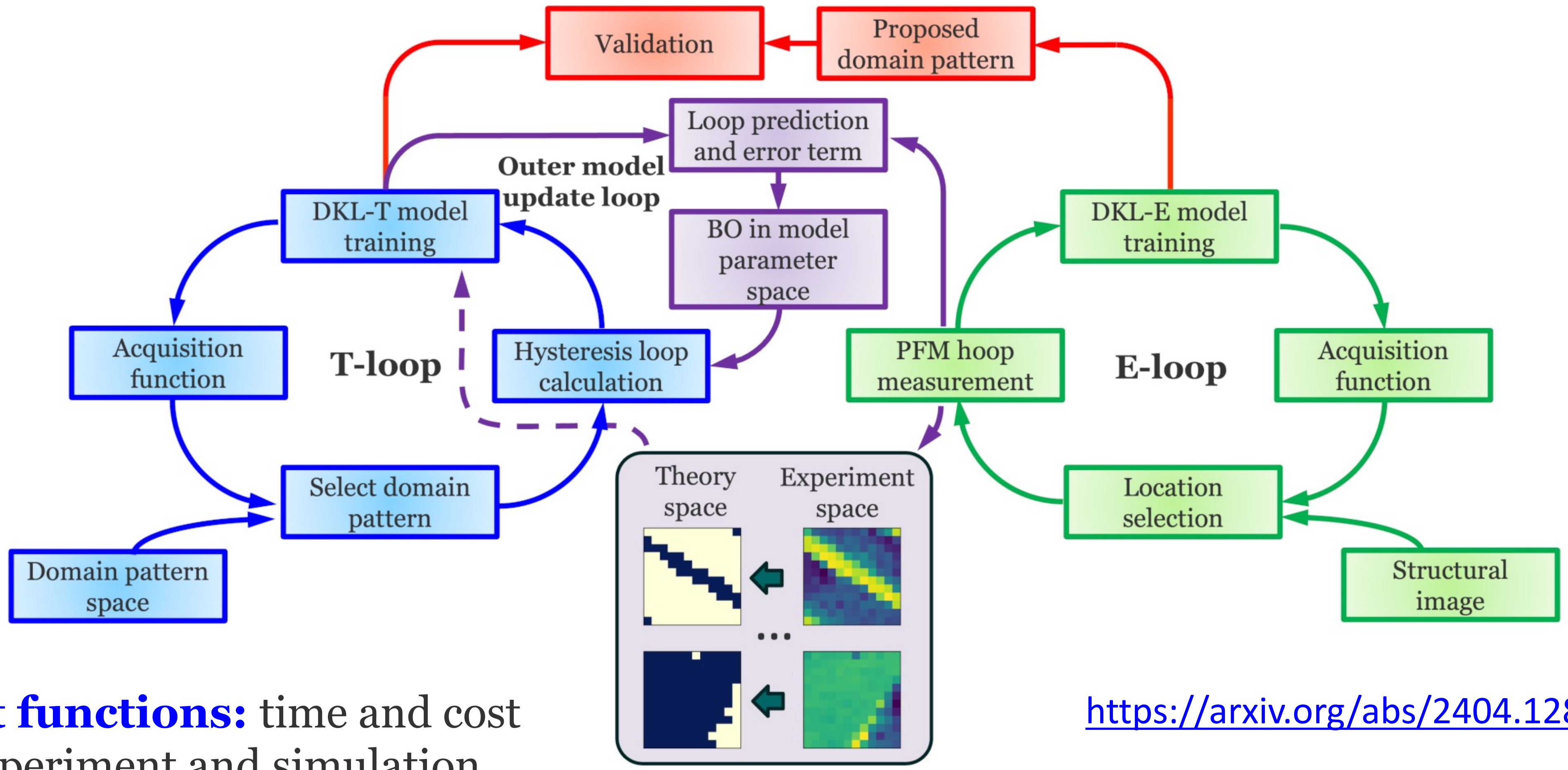
- Learned are:
  - **decoding functions**
  - spatial distribution of **latent variables**,
  - (symmetric) **correlation kernel**

- **Next challenge: causal structure in kernels**

https://arxiv.org/abs/2402.02198

Co-navigation for theory-experiment matching

- Continuous training of the surrogate model for theory (T-loop)
- Co-training surrogate model for experiment (E-loop)
- Theory update based on error function over support of interest (Outer theory update loop)



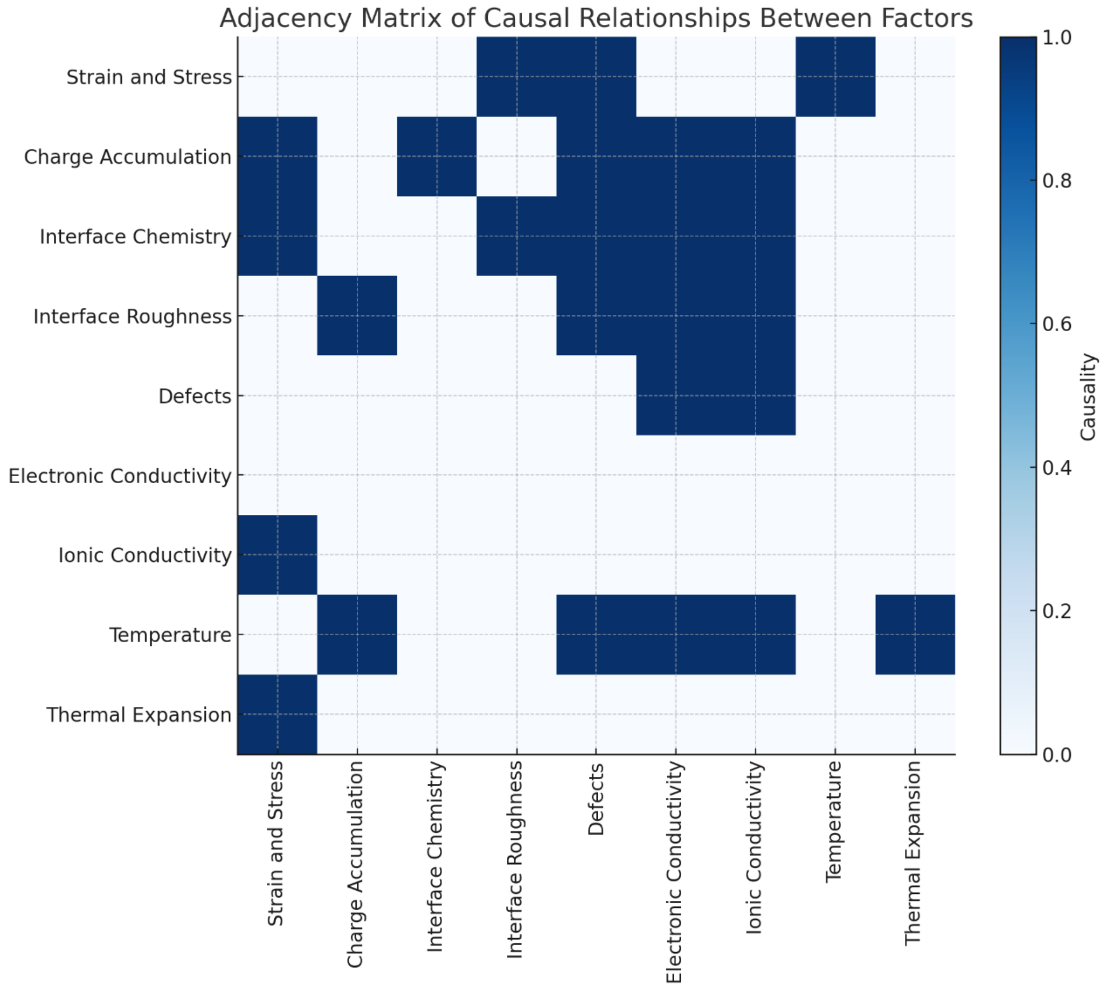
- **Cost functions:** time and cost of experiment and simulation
- **Gain:** predictive uncertainty

https://arxiv.org/abs/2404.12899

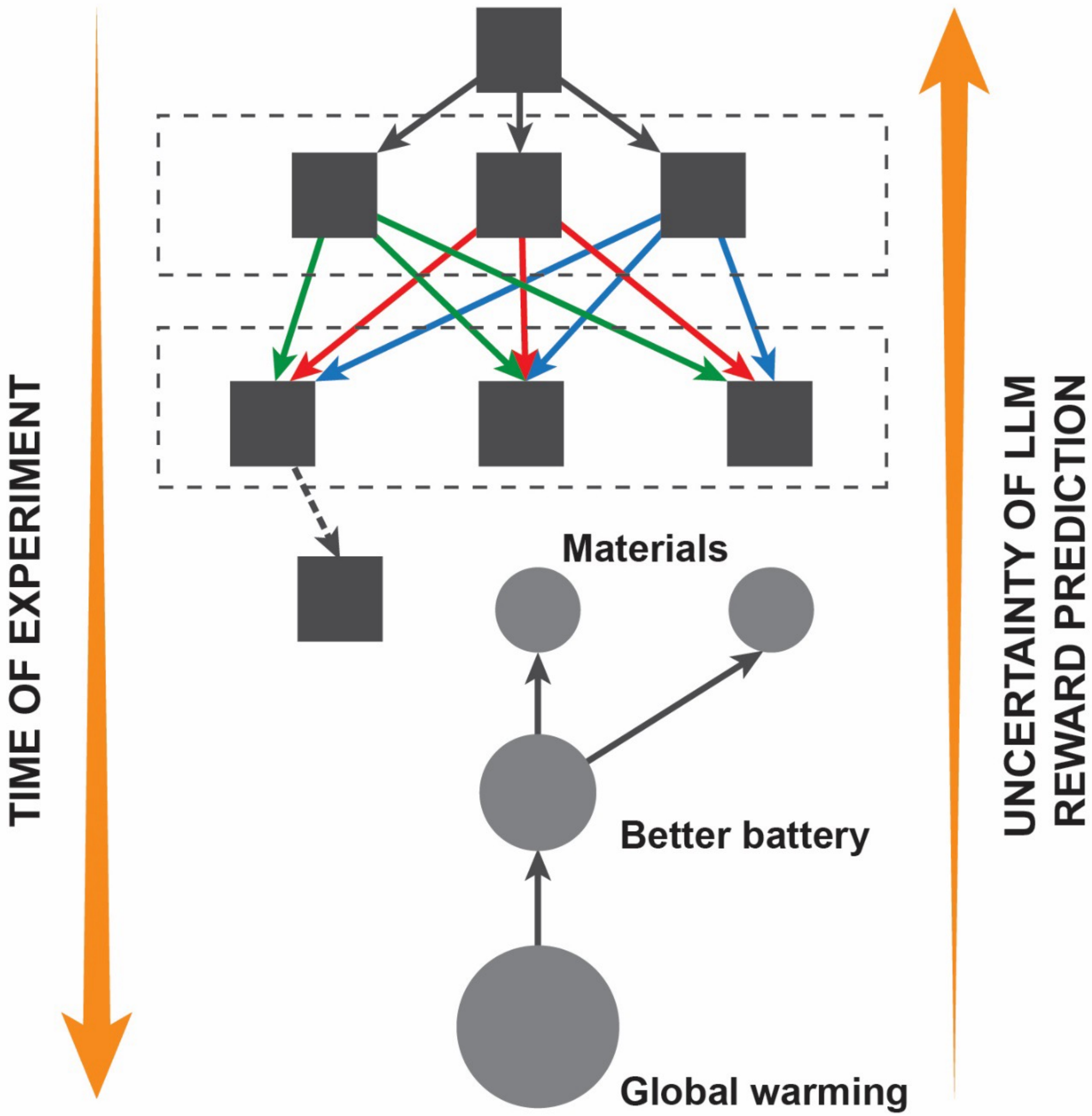
LLM Co-Scientists: Causal Discovery

```python
llm = ChatOpenAI(temperature=0, model='gpt-3.5-turbo')
tools = load_tools(["arxiv"], llm=llm)
agent = initialize_agent(tools, llm, agent=AgentType.CHAT_ZERO_SHOT_REACT_DESCRIPTION,
    handle_parsing_errors=True, verbose=False)
```

```python
def get_llm_info(llm, agent, var_1, var_2):

    out = agent(f"Does {var_1} cause {var_2} or the other way around?\
    We assume the following definition of causation:\
    if we change A, B will also change.\
    The relationship does not have to be linear or monotonic.\
    We are interested in all types of causal relationships, including\
    partial and indirect relationships, given that our definition holds.\
    ")

    print(out)

    pred = llm.predict(f'We assume the following definition of causation:\
    if we change A, B will also change.\
    Based on the following information: {out["output"]},\
    print (0,1) if {var_1} causes {var_2},\
    print (1, 0) if {var_2} causes {var_1}, print (0,0)\
    if there is no causal relationship between {var_1} and {var_2}.\
    Finally, print (-1, -1) if you don\'t know. Importantly, don\'t try to\
    make up an answer if you don\'t know.')

    print(pred)

    return pred
```



Large Language Models allow exploring body of literature via RAG to form objects (here, prior causal knowledge) that can be used to complement the data



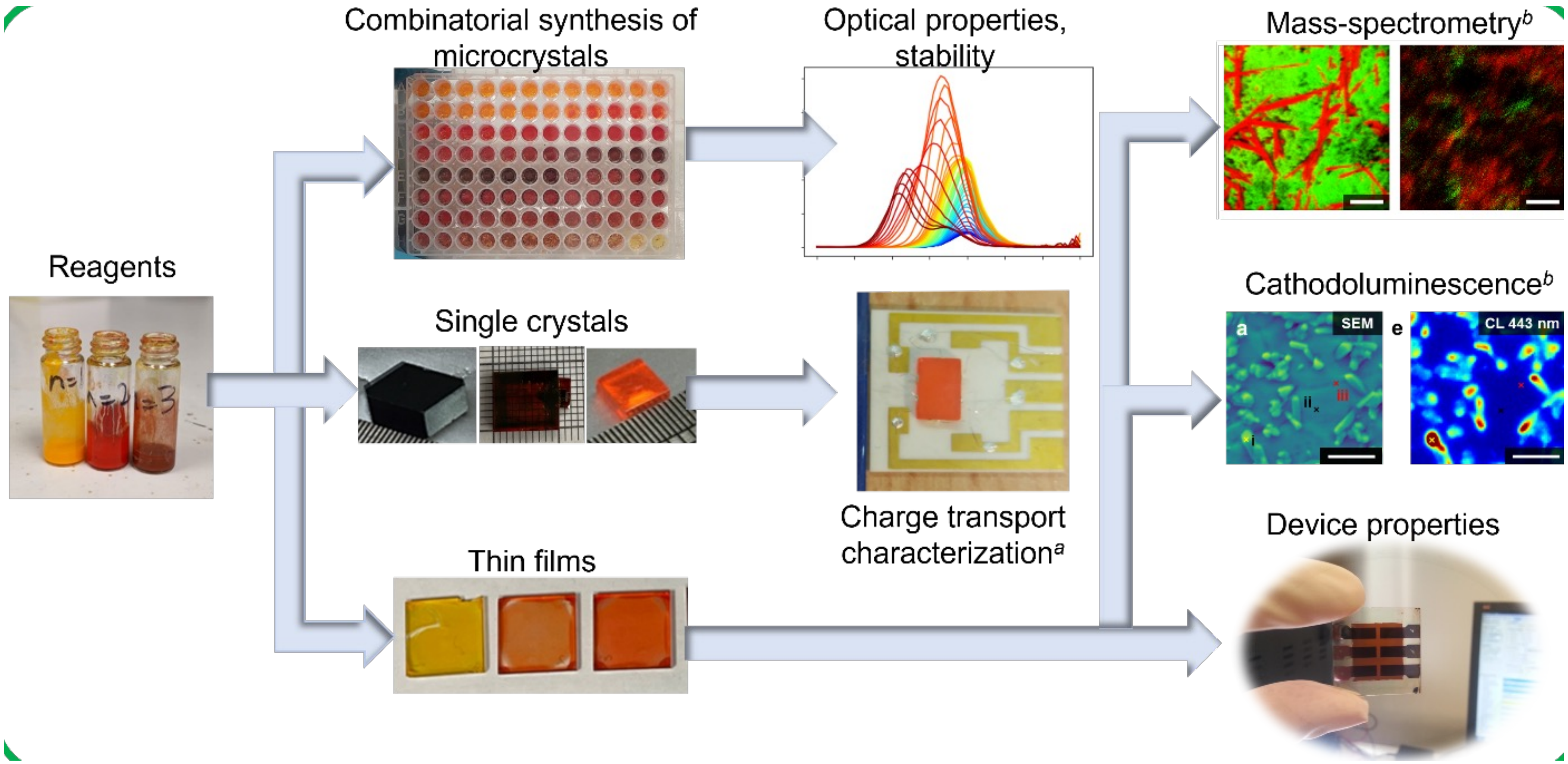
- **Big data** and associated infrastructure is useful and sometimes sufficient in well established fields with plenty of real (or perceived) downstream applications

- Automated discovery research necessitates creation of the **reward functions**, that maps discovery onto established optimization frameworks

- Large Language Models may be a viable strategy to discover probabilistic reward functions that can be refined experimentally

Thank you for attention!

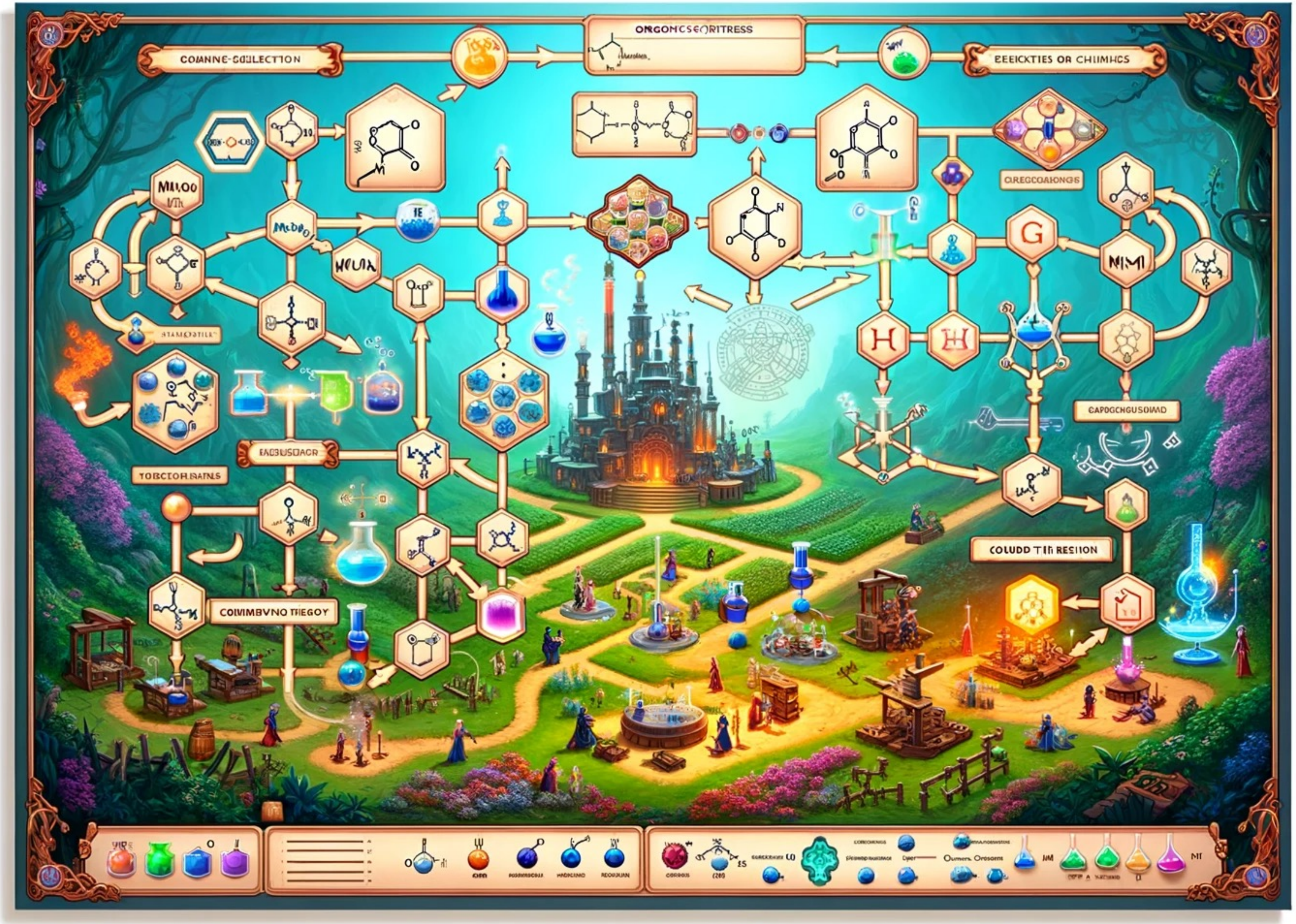
Decision science of workflows



- Multiple levels of decision making based on perceived gain, latencies, and costs
- Iterative cycles between low-cost and expensive measurements
- Learning **basic science/models** as a strategy to minimize cost and answer interventional and counterfactual questions

Homo Ludens: Gamification of SDLs

- Collaborative Environment between humans and NPCs
- Role Specialization
- Quests and Objectives
- Dynamic Interaction and Adaptation:
- Skill and Experience Growth
- Real-time Decision
- Interactive World
- Automated Tasks and Challenges
- Resource Management
- Progress Tracking and Rewards



Concluding:

- **Machine learning is great, but**
  - Requires domain expertise
  - Ease of use for deployment
  - Some ML knowledge

- **ML transition to domain areas:**
  - Reward functions
  - Workflow design
  - Hyperlanguage

- **Human in the loop – hAE**
  - Explainability
  - Interventions
  - Alignment

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