PriM: Principle-Inspired Material Discovery through Multi-Agent Collaboration

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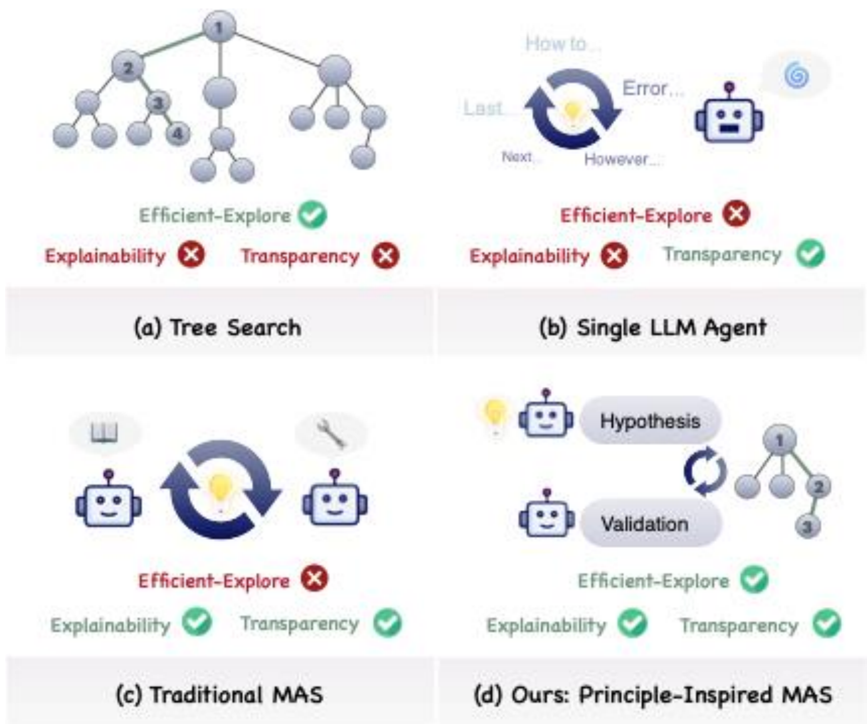
Abstract

Complex chemical space and limited knowledge scope with biases hold immense challenge for human scientists, yet in automated material discovery. Existing intelligent methods rely more on numerical computation, leading to inefficient exploration and outcomes with hard-interpretability. To bridge this gap, we introduce a principles-inspired material discovery system powered by language inferential multi-agent system (MAS), namely **PriM**. Our framework integrates automated hypothesis generation with experimental validation in a roundtable MAS, enabling systematic exploration while maintaining scientific rigor. Based on our framework, the case study of nano helix material inverse design demonstrates higher property value targeting while providing transparent reasoning pathways. This approach develops an automated-and-transparent paradigm for automated material discovery, with broad implications for rational design of functional materials.



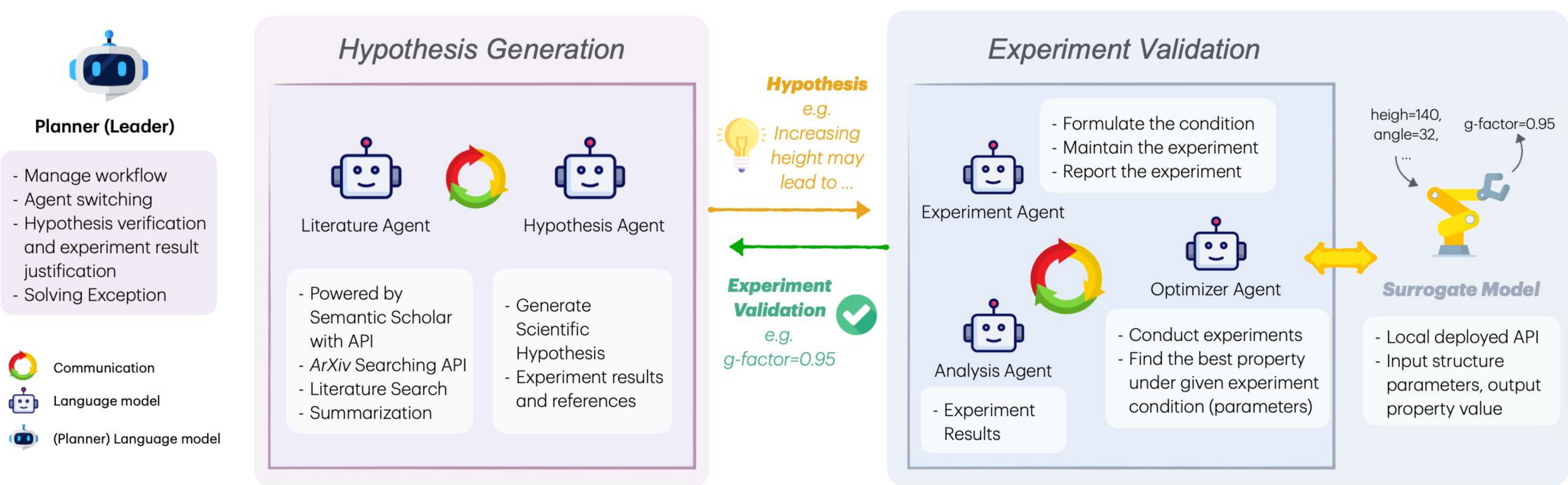
Contribution

A novel scientific discovery framework, PriM, that synergies principle-guided exploration with MAS for automated materials discovery.

Empirical validation through nano-helix material discovery demonstrates 68.6% improvement in property optimization compared to Vanilla Agent and 17.6% improvement over Vanilla MAS, highlighting the effectiveness of principle-guided exploration.

Comprehensive experiments establish advantages of PriM over existing methods.

Framework



PriM is a principle-inspired multi-agent system that alternates **hypothesis generation** and **experimental validation** under a central LLM-driven **Planner**. Before the first iteration, the user should define the research goal and constraints through a **User Proxy Agent**, after that, a **Literature Agent** is applied to retrieve literature insights. In each round, a **Hypothesis Agent** converts the knowledge into physiochemically grounded proposals, an **Experiment Agent** evaluates them in a high-fidelity surrogate virtual lab, an **Optimizer Agent** refines parameters via Monte-Carlo Tree Search, and an **Analysis Agent** distils the results into knowledge, and the **Planner** advances to the next round with the distilled knowledge.

Algorithm 1 PriM Framework for Materials Discovery

Require: Initial knowledge K_0, initial parameter set $X_0 \subset \mathcal{X}$, constraint set $\mathcal{C}$, maximum iteration M. Literature Agent L, Hypothesis Agent H, Experiment Agent E, Optimizer Agent O, Analysis Agent A and Planner $\mathcal{P}$.

Initialize $S_0 = (K_0, X_0, \varnothing)$
$\mathcal{R}_0 = \mathcal{I} = L(K_0)$ ▷ Literature insights
for $t = 1, 2, \ldots, M$ **do**
  $T_t = H(\mathcal{R}_{t-1})$ ▷ Hypothesis generation
  $D_t = E(T_t, X_{t-1})$ ▷ Experimentation
  $X^* = O(X_t, V(X_{t-1}))$ ▷ Optimization
  $R_t = A(D_t)$ ▷ Results analysis
  $X_t = X^*$ ▷ Update parameters
  $S_t = \mathcal{P}(S_{t-1}, R_t)$ ▷ State update
end for
Output: Optimal parameter $X^* \in \mathcal{X} \cap \mathcal{C}$ such that

$$X^* = \arg\max_{X \in \mathcal{X} \cap \mathcal{C}} f(X).$$

The PriM framework is formalized in Algorithm 1, and Theorem 1 proves the theoretical convergence of PriM.

Theorem 1. *Let the parameter space $\mathcal{X}$ be compact and let $f : \mathcal{X} \to \mathcal{Y}$ be continuous. Under the update dynamics of PriM, the sequence $\{f(X_t)\}_{t=1}^{\infty}$ is monotonically non-decreasing and converges to the optimal value $f(X^*)$. Specifically,*

$$\lim_{t\to\infty} |f(X_t) - f(X^*)| = 0.$$

Methodology summary: Guided by fundamental physicochemical principles generated by the Hypothesis Agent, PriM strategically constrains partial experimental conditions to allow numerical optimization in a reduced property space, thereby accelerating the identification of target material structures.

Baselines and Metrics

Apart from numerical search algorithms, we consider the following baselines:

1. **Vanilla Agent**: Single optimization agent with naïve hypothesis generator.
2. **Vanilla MAS**: The full roundtable of PriM *except* the Hypothesis Agent.
3. **AccelMat** [1]: A recent MAS for material discovery that focuses on hypothesis generation.
4. **MASTER** [2]: An MAS that augments MCTS with LLM-based heuristics.

Metrics:

1. Optimal material property value (real value μ and proportional value μ_{rel});
2. Normalized exploration rate (ε); 3. Convergence iterations.

Results and Analysis

PriM achieves competitive material property values (μ) while maintaining scientific rationality.

Table 1: Comparison of PriM with Baselines Methods (mean ± std)

Method	Optim. Val. (μ)	μ (%)	Exp. Rate (ϵ)	Conv. Iter.
BO [11]	1.334 ± 0.07	90.7	0.94 ± 0.05	84.8 ± 12.79
DQN [26]	1.157 ± 0.17	78.6	0.74 ± 0.13	24.7 ± 7.56
MCTS [5]	1.102 ± 0.05	74.9	1.22 ± 0.02	68.3 ± 21.44
Vanilla Agent	0.644 ± 0.05	43.8	0.65 ± 0.02	9.20 ± 2.56
Vanilla MAS	0.923 ± 0.17	62.7	0.35 ± 0.11	65.40 ± 18.91
AccelMat [20]	0.625 ± 0.24	42.5	0.54 ± 0.30	25.00 ± 17.36
MASTER [13]	0.440 ± 0.12	29.9	0.39 ± 0.19	16.00 ± 4.65
PriM (Ours)	$\mathbf{1.086} \pm 0.05$	**73.8**	0.45 ± 0.07	85.50 ± 8.58

Table 1 validates the performance of PriM through comparison with other baseline methods.

Table 2: Component Ablation (mean ± std)

Setting	Optim. Val. (μ)	μ (%)	Exp. Rate (ϵ)	Conv. Iter.
PriM	$\mathbf{1.086} \pm 0.05$	**73.8**	0.45 ± 0.07	85.50 ± 8.58
w/o Hypothesis Agent	0.923 ± 0.17	62.7	0.35 ± 0.11	65.40 ± 18.91
w/o Literature Agent	0.923 ± 0.11	62.7	0.41 ± 0.06	83.00 ± 14.98

Table 2 proves the contributions of literature agent and hypothesis agent through component ablation.

Table 3: Hyperparameter Ablation (mean ± std)

(a) Difference in MCTS Iterations

Setting	Optim. Val. (μ)	μ (%)	Exp. Rate	Conv. Iter.
10 Iterations	0.856±0.08	58.1	0.48±0.08	43.0±5.4
20 Iterations	1.086±0.05	73.8	0.45±0.07	85.5±8.6
30 Iterations	1.100±0.12	74.8	0.69±0.03	121.8±26.2
40 Iterations	1.024±0.07	69.6	0.68±0.07	135.0±46.7
50 Iterations	1.041±0.10	70.7	0.46±0.08	203.4±37.2

(b) Difference in Optimizer

Setting	Optim. Val. (μ)	μ (%)	Exp. Rate	Conv. Iter.
MCTS [5]	**1.086**±0.05	**73.8**	0.45±0.07	85.5±8.6
BO [11]	0.966±0.05	65.7	0.62±0.04	N/A
DQN [26]	0.472±0.25	32.1	0.52±0.08	N/A

(c) Difference in Language Model

Setting	Optim. Val. (μ)	μ (%)	Exp. Rate	Conv. Iter.
GPT-4o [1]	**1.086**±0.05	**73.8**	0.45±0.07	85.5±8.6
Qwen 2.5 [40]	0.826±0.08	56.1	0.33±0.04	82.0±11.7
Gemini Pro [33]	0.868±0.11	59.0	0.59±0.06	86.0±6.5

Case Study

- **Research objective**: *Find the structural parameters corresponding to the strongest chirality (g-factor characteristics) in the nano helix material system.*
- **Research constraint**: *Explicitly show the underlying physicochemical principles regarding the structure and property relationships.*
- **Literature insights**: *Quantum-Chemical Study of the Photophysical Behavior of Mesogenic Europium(III) Complexes with β-Diketones and Lewis Bases* with its summarization establish that **coordination polyhedra govern optical properties** in complex materials.
- **Hypothesis generation**: *By optimizing the helix radius to an initial value of 55 (within the range of 20 to 90), the nano helices material system will exhibit the strongest chirality (g-factor characteristics), as the helix radius significantly influences the coordination polyhedra and optical properties, aligning with the physicochemical principles of structure-property relationships highlighted in the literature.*

We summarize the principle in hypothesis generation over iterations:

- **Iter. 1**: *Helix radius* governs coordination geometry and optical anisotropy, critically tuning chirality in accordance with structure-property principles.
- **Iter. 2**: *Helix radius* and *the number of turns* jointly regulate structural symmetry and ligand field effects, driving chiral enhancement through coordinated geometry.
- **Iter. 3**: Interplay among *helix radius*, *pitch*, and *the number of turns* modulates helical symmetry and nonlinear optical response, enabling peak chiral performance.
- **Iter. 4-6**: Synergistic tuning of *helix radius*, *pitch*, and *fiber* geometry shapes optical anisotropy and structural rigidity, reinforcing chiral amplification.
- **Iter. 7-8**: Coordinated modulation of *helix radius*, *pitch* and *the number of turns* tunes chirality via structural asymmetry, guided by CD spectra and bio-inspired design.

g-factor evolution over iterations: 0.418 → 0.625 → 0.706 → 0.95 → 0.974.

Impact Statement

To the best of our knowledge, this work represents the first exploration of **principle-driven materials discovery (PMD)** using language models. Our PriM framework demonstrates a significant advancement in automated materials discovery by **integrating scientific principles into exploration process** through multi-agent collaboration. By embedding physicochemical principles into hypothesis generation and experimental validation, PriM not only achieves superior performance in identifying optimal material properties but also maintains interpretability throughout the discovery process. This approach bridges the gap between **black-box optimization** and **scientific understanding**, paving the way for AI-assisted discovery that remains grounded in established scientific methodology.

Limitations and Future Work

The reliance on LLM inference introduces potential biases or hallucinated correlations between structure variables and property values. Though PriM includes verification mechanisms, further work is needed to quantify and mitigate these biases. In addition, we plan to incorporate advanced reasoning mechanisms that refines hypothesis selection through structured causal inference, this may potentially improve exploration efficiency and interpretability. We also aim to extend PriM to more complex scientific systems where mechanistic understanding is limited, where principle guided exploration could be particularly valuable.

References

[1] Kumbhar, S., Mishra, V., Coutinho, K., Handa, D., Iquebal, A., & Baral, C. (2025). Hypothesis Generation for Materials Discovery and Design Using Goal-Driven and Constraint-Guided LLM Agents. *arXiv preprint arXiv:2501.13299*.

[2] Gan, B., Zhao, Y., Zhang, T., Huang, J., Li, Y., Teo, S. X., ... & Shi, W. (2025). MASTER: A Multi-Agent System with LLM Specialized MCTS. *arXiv preprint arXiv:2501.14304*.