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Comparison of FePt high-density magnetic recording media development between large language models and experimental experts: do LLMs recommend fluoride as a segregant?

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ABSTRACT

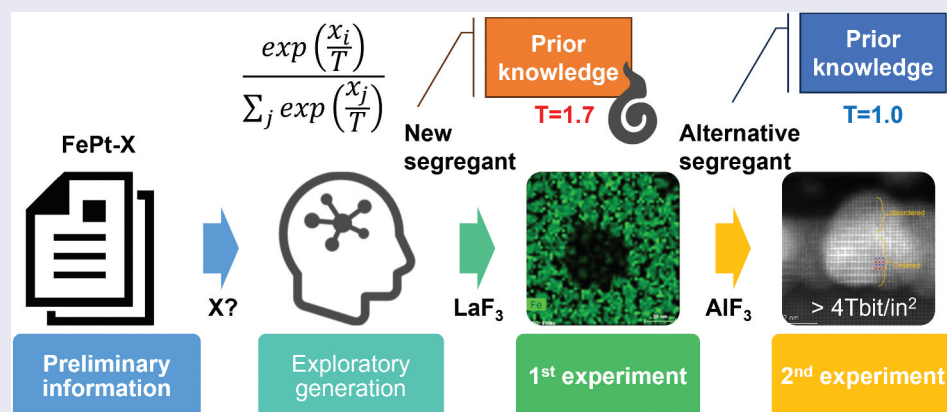
We investigated segregant development for FePt high-density magnetic recording media using large language models (LLMs) and identified new fluoride-based materials. By increasing the temperature of the softmax function to select probable vocabularies, we successfully incorporated the prior knowledge involved in LLM training into the material selection process (exploratory generation) and identified LaF₃ as an optimal material. Meanwhile, we independently conducted sputtering deposition of FePt-LaF₃ nanogranular samples and verified whether the LLM could reproduce the results. The inhomogeneity in the surface chemical composition of FePt-LaF₃ in the non-equilibrium state of sputtering was also reproduced by the LLM, leading to the identification of AlF₃ as an alternative segregant. Structures and magnetic properties strongly dependent on the sputtering apparatus cannot be reproduced by simple LLM predictions, necessitating interactive data exchange between physical and cyber experiments. However, phenomena that can occur in physical experiments are almost accurately reproduced by LLM, demonstrating the usefulness of LLM predictions in material development. LLMs that generate responses from the most likely vocabulary tend to select well-known materials. To identify unknown materials as candidates, techniques such as exploratory generation or inputting unexpected results are required.

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IMPACT STATEMENT



By using prior knowledge embedded in LLM, fluorides were discovered as a segregant for FePt nanogranular films. Using AlF₃, FePt nanoparticles (particle size: 3.8 nm, equivalent to > 4 Tbit/in²) were realized.

1. Introduction

1.1. Background of this study

Large language models (LLMs) are now being used in materials development [1–3]. However, because LLMs are basically designed to generate answers from questions, it is currently unknown whether they can

autonomously explore new materials. Conceptually, it should be possible to extract guidance from questions and answers (Q&A) and then semi-autonomously derive the best solution. However, at least the following points need to first be verified.

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- (1) Is it possible to predict unknown materials from the pre-trained data of the LLM, i.e. prior knowledge (Novelty of prediction)?
- (2) Does the LLM have sufficient background knowledge about the target materials (Validity of prediction)?
- (3) If material prediction is possible, how well does it match experimental facts (Probability of prediction)?

To clarify these three issues, in this study, we performed the following.

(1) Novelty of prediction

Considering that the pre-training (prior knowledge) resources of an LLM are already known information, strikingly new facts may not emerge. However, it is possible to combine existing information and incorporate themes that have not yet been discussed in a specific field, thereby potentially allowing for predicting new materials. In this paper, we refer to the process of generating novelty in a narrow, specialized field from a broad range of existing information as ‘exploratory generation’, and we attempted to predict previously unknown materials in the specific field using exploratory generation.

(2) Validity of prediction

As will be discussed in detail later, benchmark Q&A sets that serve as evaluation metrics for science and mathematics are well known. However, when considering new material development as a task, it is necessary to confirm that LLM has knowledge of basic physical properties and the necessary background for material prediction. Because an LLM generates the most plausible words in natural language processing, it is possible to generate statements that contradict physical truths. Therefore, we verified that predictions containing contradictions can be avoided by confirming quantitative physical constants and their background knowledge.

(3) Probability of prediction

We verify the capabilities of the LLM by identifying gaps between independent predictions and experimental facts, rather than directly inputting experimental results into the LLM. In actual material development, it may be possible to iteratively improve prediction accuracy by sequentially communicating experimental results, or to optimize processes under models such as Bayesian optimization [4]. However, in this study, we focused on verifying the probability of LLM predictions by examining how well the LLM can trace the experimental results conducted by experts and identify where unpredictable aspects emerge.

1.2. Background of physical experiments

In this study, the target of the LLM for the exploratory generation of new materials and prediction of what

occurs during their fabrication process was FePt-X nanogranular films, which are attracting increasing attention as next-generation high-density magnetic recording media. Here, X is a non-magnetic segregant that separates FePt magnetic nanoparticles. FePt shows large magnetocrystalline anisotropy in a chemically ordered L1₀ phase [5]; therefore, the effect of X on this nanostructure is the main topic of this study. The reader is referred to [6–8] for background regarding the development of FePt-X as well as the current state of the art. FePt-X nanogranular films are deposited using an ultra-high vacuum sputtering system, and their shape, chemical composition distribution, crystallinity, and magnetic properties are evaluated.

The key points in the practical development of magnetic recording media using FePt-X are as follows.

- Minimization of magnetic particle size (<5 nm) by introducing a new X
- Stability of X; that is, minimizing the decomposition of X, its diffusion into FePt, and its reaction
- Simultaneous realization of small structure and high magnetic properties

It should be noted that a specific model is necessary to predict magnetic properties from nanoparticle shapes. Without such a model, it is difficult to predict magnetic properties using an LLM for actual samples involving decomposition, diffusion, and the reaction of X. In this study, the aim is to verify material exploration procedures using an LLM rather than model development, and magnetic properties are used as indirect indices to verify the validity of structural and chemical predictions made by the LLM. Fortunately, indirect indicators are sufficiently reasonable because the magnetically favorable structure (ordered L1₀ phase in FePt without chemical contamination) and the challenges in crystal growth for improving magnetic properties (competition between separation within the horizontal plane and columnar growth) are well understood [8,9].

1.3. Background of cyber experiments

Gemini 2.5 Flash and Pro, which were used in this study, are modules developed by Google that are characterized by high reasoning performance through the modeling of human thought processes. Table 1 summarizes the evaluation results using publicly available

Table 1. Summary of evaluation results of Gemini 2.5 using publicly available benchmarks.

Benchmark	Score	Target field
Science GPQA diamond	84.0%	Scientific [10]
Mathematics AIME 2025	86.7%	Mathematics [11]
Mathematics AIME 2024	92.0%	Mathematics [11]

benchmarks [10]. The benchmark scores for science [11] and mathematics [12] are high, indicating sufficient reasoning ability in logic-based content. Furthermore, Gemini was selected because it is expected to provide sufficient predictive capability compared to other models [13]. Although various evaluation criteria for models have been proposed, such as more complex problem settings [14] and robustness against noise [15], it should be noted that these benchmarks are based on the assumption that the solution method is already known and therefore cannot be used as a direct indicator of applicability to the development of unknown materials. However, if an LLM can reproduce the human thought process, it should be possible to verify the extent to which research plans can be formulated and new facts can be discovered by comparing them with experts.

In addition, Gemini clearly demonstrates the thought process of problem solving and offers a high level of transparency. This is convenient for science, which emphasizes explainability, and serves as a basis for human judgment on whether to proceed to physical experiments in the event of competition. Furthermore, because it can handle 1 million tokens [16], it is possible to conduct comprehensive discussions. However, in this study, we focus on a specific issue (exploration of new segments) to evaluate the material exploration capabilities of the LLM within a very limited field. The scope for exploring materials is normally vast, and various factors must be taken into consideration. Attempting to balance all these factors results in only well-known, safe materials being proposed, despite the abundance of knowledge available. To avoid such non-challenging predictions, it is necessary to focus on specific issues, reduce the number of factors that need to be considered, and create a situation in which innovative answers can be derived.

2. Cyber experiment methods

Verification of the novelty of prediction began with the LLM predicting the target segregant X. Then, once X is determined, physical and cyber experiments will not interfere with each other even if they handle the same X. This section describes the cyber experiment method.

For example, using retrieval-augmented generation (RAG) [17], it is possible to search for materials in a specific theme. When developing X as a theme, RAG divides related papers into chunks, stores them in a database in advance, extracts chunks highly relevant to X, and generates answers. Therefore, this method is completely interpolative and is good for researching known materials. However, it does not lead to the discovery of new

materials. What we need for the ‘exploratory generation’ of new X is to

- share the minimum amount of known information (preliminary information) necessary for the LLM to understand the task and
- explore X from prior knowledge.

In the case of RAG, the more relevant information there is, the richer the search results are expected to be. However, the results are clearly different from what is required in the current study. The key is how to extract information ‘other than’ what is already available. LLMs use a softmax function to convert the absolute scores (logits) of the vocabulary to be selected into a relative probability distribution. The softmax function is expressed by the following equation:

$$\text{Softmax}(x_i) = \frac{e^{x_i}}{\sum_j e^{x_j}}$$

By introducing temperature T for logit variable transformation from x_i and x_j to x_i/T and x_j/T , it is clear that as T increases, the probability distribution widens, and answers containing wider candidates of vocabulary (including X) are generated. In this study, we explore new materials by increasing T . Once a material is discovered, T was set to the default value of $T=1.0$ to verify the validity and probability of the prediction to obtain more solid evidence. When exploring new materials, it is easy to understand that the range of preliminary information provided to help define the problem should be limited so that the LLM does not propose known materials. In this study, we selected seven papers [18–24] for the keyword ‘phase separation’, four papers [25–28] for ‘grain density’, five papers [29–33] for ‘surface free energy’, and one paper on the ordered $L1_0$ phase at low temperatures [34] from previous reports on FePt as preliminary information. The segregants discussed in these papers are primarily C [18–21,23–26,33], SiO_2 [18,19], TiO_2 [18], and Cr_2O_3 [27,28] and other metal oxides [19,22,30]. Although the fluorides to be discussed in the current paper are not included, the requirements for segregants and the basic information necessary for prediction are sufficiently covered. By conducting several Q&A sessions for each of the above keywords in advance, we clarified the task’s intent and shared the desired direction for prediction with the LLM. The sequence of questions conducted to arrive at the conclusions presented in this paper is summarized in [Appendix A](#). The section concerning segregant prediction is detailed in [Appendix A \(1\)](#).

3. Cyber experiments (I)

3.1. Novelty of prediction

Gemini 2.5 Flash was used to compare $T = 1.0$ and 1.7 . First, papers containing preliminary information were fed into Gemini to summarize the current state of FePt development from the perspective of phase separation, requirements and challenges, and answers to the next steps in development, confirming that the LLM had a sufficient understanding of the details of the state of the art (c.f., Appendix A (1)). Then, a list of promising X was generated. The LLM predictions for X, excluding materials with complex layer structures, are summarized in Table 2. $T = 1.0$ afforded specific and well-known materials, while $T = 1.7$ provided many materials with general expression showing examples. To confirm novelty, we investigated the number of FePt-related paper titles containing oxides, nitrides, borides, and fluorides using the following search query:

allintitle: FePt h-BN (Example of a search query for hexagonal boron nitride as a segregant) A histogram of relevant papers for each material is summarized in Figure 1. The majority of papers are related to oxides and nitrides, as listed in Table 2 for $T = 1.0$, indicating that these materials are not novel. Furthermore, no papers related to borides and fluorides were found, suggesting that exploratory generation works well for the LLM with $T = 1.7$.

Appendix B shows the answers predicted for fluorides by the LLM with $T = 1.7$. The reasons for selecting fluorides included the stability of ionic compounds and their low reactivity with Fe and Pt. However, low interface energy was pointed out as a potential issue, as it could lead to significant island growth and hinder columnar growth. In addition, concerns were raised about the competition between the high temperatures required for the ordering of FePt and the low process temperature to suppress fluoride decomposition. Thus, the proposal of a high-risk material indicates that the softmax function is producing the expected effect. The exploratory generation is considered to depend not only on T but also on the preliminary information. Details will be discussed in Section 7.

Another novel material class is metal borides. However, this is simply because they are hard and heat-resistant, and there is insufficient evidence to demonstrate the advantages of these materials.

Table 2. Summary of LLM predictions for X.

	T = 1.0	T = 1.7
Oxide	Y ₂ O ₃ , HfO ₂ , TaO ₅	Complex oxides
Nitride	TiN, Si ₃ N ₄	e.g., TiN, ZrN, HfN, Si ₃ N ₄ , BN
Boride		e.g., TiB ₂ , ZrB ₂ , TaB ₂ , HfB ₂
Fluoride		e.g., CaF ₂ , SrF ₂ , LaF ₃

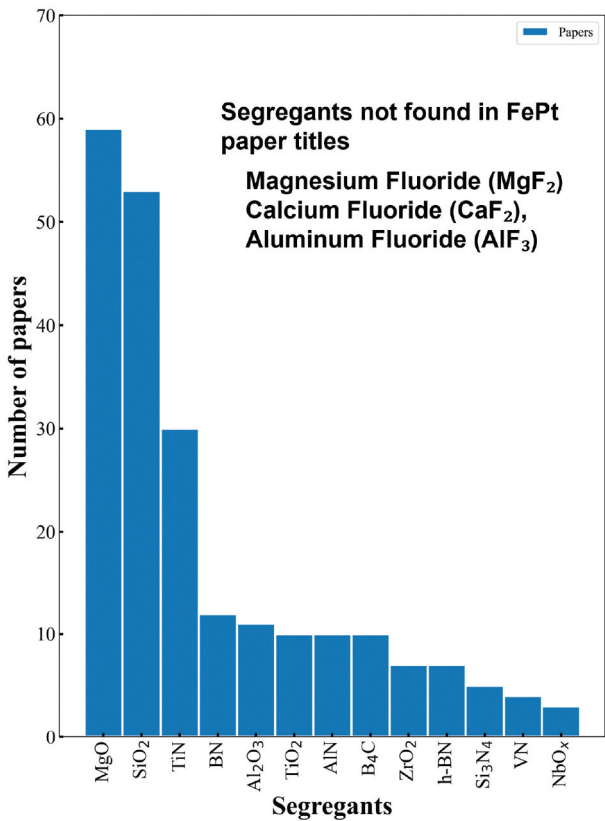


Figure 1. Number of FePt-related papers in Google Scholar with titles containing oxides, nitrides, metal borides, or fluorides.

Therefore, we decided to focus on fluorides. As can be understood from the fact that no predictions were made for fluorides with $T = 1.0$, tuning T can be used as a method of adjusting novelty and reliability. Considering the influence of prior knowledge and the inherent imperfections of LLMs, it may be appropriate to leave room for the human selection of LLM responses for collaborative material development by LLMs and experts. Otherwise, there is a risk of becoming locally trapped in systems with high uncertainty.

We have summarized the answers from Gemini 2.5 Pro $T = 1.0$, which provided more reliable results, in Appendix C. The reasons for not listing fluorides include the fact that fluorine decomposes during sputtering, and due to its high reactivity, it may corrode FePt. In addition, there is limited research on its interface energy, making it difficult to determine whether columnar growth will occur. As expected, this contradicts the claims made by Gemini 2.5 Flash with $T = 1.7$, indicating that there is a boundary where fluoride compounds are recommended or not. Clearly, fluoride compounds present numerous challenging factors, and as will be discussed later, film formation inherently involves many barriers. Therefore, we sought supporting evidence for making fluoride compounds candidate materials, conducted extensive Q&A sessions with the LLM, and ultimately became confident in their

validity. The types and number of questions, along with an overview of representative content, are summarized in [Appendix A \(2\)](#).

3.2. Validity of prediction

As mentioned in [Section 1.3](#), Gemini's logical thinking appears to be at a high level. In such a situation, the validity of new material candidates is thought to heavily depend on the reliability of the data used for consideration. Here, we verified the relevant physical property information that Gemini has as prior knowledge. In the following discussion, we use Gemini 2.5 Pro with $T = 1.0$ to summarize the values obtained from reliable information. Although this verification is specifically tailored to the development of FePt segregants, it can be considered a similar approach to a benchmark for predicting physical property values [35].

[Figure 2](#) shows a comparison of the Gibbs free energy of formation (ΔG^0_f) values obtained from literature [36] and those provided by the LLM. The values obtained by the LLM are slightly larger, but they agree very well with literature.

The LLM understands the physical meaning of Gibbs free energy:

“Gibbs Free Energy of Formation (ΔG^0_f):

A large negative ΔG^0_f value indicates that the compound is much more stable than its constituent elements in their standard states, making it less likely to decompose or react.”

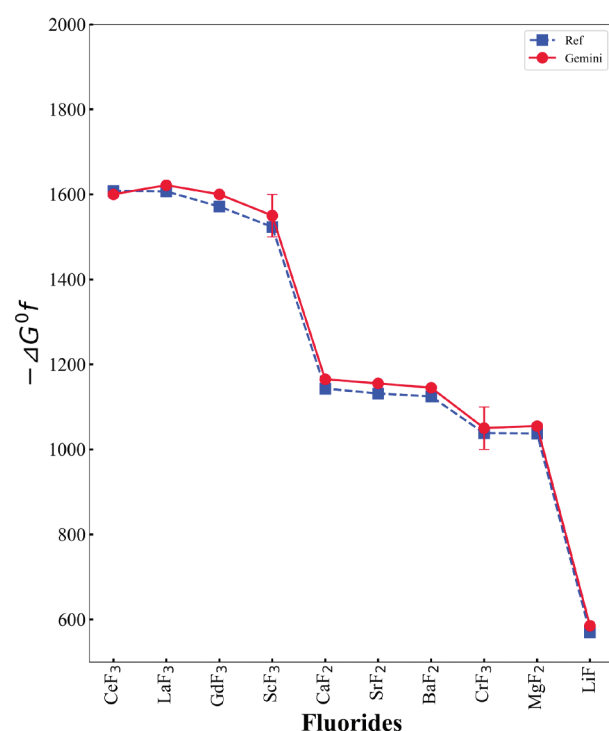


Figure 2. Comparison of Gibbs free energy of formation, ΔG^0_f obtained from literature and those provided by the LLM.

On the other hand, the LLM pointed out that Gibbs free energy is only a partial evaluation when selecting fluoride as a segregant, as follows:

“Important Notes When Considering These:

Kinetic Factors & Practicality:

Volatility/Sublimation: Even with a negative ΔG^0_f , if the vapor pressure becomes significant at deposition temperatures, the material won't work as a stable segregant. This is a major concern for compounds like ZrF₄ and HfF₄.

Fluorine Reactivity: The risk of fluorine release and reaction remains a key practical hurdle.

interface energy with FePt: High thermodynamic stability doesn't guarantee the correct interface energy for optimal phase separation.

Sputterability and Film Formation: The ability to deposit these as thin, conformal, and stoichiometric films.

These suggestions expand the list based on the thermodynamic stability criterion. Each would require careful experimental evaluation for all the other critical properties needed for an effective FePt segregant.”

From this, we can see that the LLM is capable of demonstrating reasoning and objectively reporting when the reasoning is not sufficiently valid.

To identify a better metal fluoride for physical experiments, we identified another physical constant using the LLM. In particular, controlling the F reactivity to maintain the stoichiometry of fluorides may compete with the diffusivity of metals in the segregant. That is, if metal diffusion is fast, the possibility of incorporation into FePt increases. The ionic radius serves as an index of diffusion, and we verified whether the LLM possesses this information. [Table 3](#) shows a comparison of the Shannon ionic radii of various metals for a coordination number (CN) of six obtained from reference [37] and those obtained using the LLM. The values of both sources are almost identical, and their order is also identical except for Yb³⁺. In fact, Gemini accurately expresses the utility of the ionic radius as follows:

“Reasoning for Considering Larger Ionic Radii:

Slower Diffusion: Larger ions generally have higher activation energies for diffusion through a lattice because they cause more distortion when moving between sites or through interstitial positions. If the metallic element from the fluoride segregant were to try and diffuse into the FePt lattice, a larger size would make this process kinetically less favorable.

Table 3. Ion radius (pm) of each metal for CN=6.

	Shannon	Gemini
Ba ²⁺	135	135
Sr ²⁺	118	118
La ³⁺	103.2	103
Yb ³⁺	102	86.8
Ca ²⁺	100	100
Gd ³⁺	93.8	93.8

Lattice Strain/Solubility: A significantly larger ion would create substantial strain if it tried to substitute for Fe or Pt atoms in the $L1_0$ lattice, leading to very low solid solubility. This reinforces the immiscibility criterion.”

Considering Figure 2 and Table 3 together, LaF_3 can be concluded to be the most promising candidate. Specifically, LaF_3 is a stable metal fluoride with a stability (ΔG^0_f) exceeding -1600 kJ/mol, and the ionic radius of La^{3+} is large, exceeding 100 pm for CN = 6, which is expected to suppress diffusion into FePt.

3.3. Probability of prediction

As discussed in Section 3.2, LaF_3 was confirmed as the segregant to be verified. As will be described later, the probability of the prediction will be demonstrated by physical experiments. Here, we used the LLM to predict what is likely to occur in the reaction with fluorine. As shown in Appendix D, the following reactions cannot occur, i.e. once chemically stable LaF_3 is formed, it will not decompose and fluorinate Fe or Pt as follows:

Reaction 1: $\text{LaF}_3(\text{s}) + \text{Fe}(\text{s}) \rightarrow \text{La}(\text{s}) + \text{FeF}_3(\text{s})$

Reaction 2: $2\text{LaF}_3(\text{s}) + \text{Pt}(\text{s}) \rightarrow 2\text{La}(\text{s}) + \text{PtF}_6(\text{s/g})$.

However, if excessive fluorine is generated in a non-equilibrium state such as in a high vacuum sputtering system, it is possible that volatile Fe or Pt fluorides will be formed. The LLM predicts that such a situation is possible, as shown in Appendix E. In the statements of ‘1. Preferential Sputtering or Resputtering of Lanthanum’ and ‘2. Plasma-Induced Dissociation of LaF_3 and Differential Sticking/Reaction of Species’, (1) differences in the sputtering rates of La and F, or the resputtering of La from the sample by sputtering ions and (2) the generation of non-stoichiometric decomposition species due to gas-phase reactions in the plasma, and chemical composition imbalances caused by differences in surface sticking coefficients and reaction coefficients are predicted to result in the formation of excess fluorine. This is also a very reasonable prediction, and while it is not particularly novel, it is clear that the LLM has general knowledge about plasma reactions.

The next step is to consider the reaction of excess fluorine with FePt and the possibility of evaporation. Table 4 shows the evaporation predictions for Fe and Pt fluorides at temperatures ranging from 400°C to 600°C using the LLM. Based on this prediction, Pt is expected to evaporate as PtF_4 or PtF_6 , while Fe may evaporate as FeF_3 . As shown in Appendix F, the LLM does not have vapor pressure data. Furthermore, because processes where

growth and evaporation occur simultaneously are complex and lack generalizability, clear reaction or evaporation predictions cannot be made. For such complex systems, we believe that physical experiments should be relied upon. In this paper, we refer to situations where predictions based on general prior knowledge are impossible as ‘Prediction of non-general events’.

Nevertheless, if a stoichiometric equilibrium is established, fluorides are extremely stable, but the generation of excess fluorine and the accompanying adsorption and desorption of various fluorides in a non-equilibrium state are expected to cause inhomogeneity in the chemical composition and size of FePt- LaF_3 on the surface. This is the conclusion of the cyber prediction presented here.

4. Physical experiments (LaF_3 segregant)

4.1. Experimental methods

Here, we discuss physical experiments using LaF_3 predicted by the LLM as a suitable segregant. The probability of predictions will be verified by comparing the physical experiments described below with the cyber experiments described in Section 3. The physical experimental methods are as follows.

The samples were fabricated by an ultra-high vacuum magnetron co-sputtering system with a base pressure of $\sim 10^{-7}$ Pa. The film consisted of a stack of FePt (0.5 nm, 400°C)/FePt-30 vol% LaF_3 (2.0 nm, 400°C)/C (5 nm, room temperature (RT)). FePt acted as the nucleation layer to increase the grain density. The 5 nm-thick C capping layer prevents surface damage. The substrate was a MgO (001) single crystal substrate. Before the deposition, MgO substrates were annealed at 650°C for 30 min.

Transmission electron microscopy (TEM) coupled with energy-dispersive X-ray spectroscopy (EDS) was performed using a TITAN G2 80–200 TEM. Magnetic hysteresis measurements were conducted using a superconducting quantum interference device (SQUID) equipped with a vibrating sample magnetometer (VSM) (MPMS, Quantum Design) with an applied magnetic field of up to ± 7 T. Cross-sectional TEM lamellae were prepared using the focused ion beam (FIB) lift-out technique (FEI Helios Nanolab G5). In-plane TEM samples were prepared by the etching method. The phase, crystal structure, and texture were investigated using X-ray diffraction (XRD) (Rigaku SmartLab).

Table 4. Evaporation prediction of Fe and Pt fluorides in the temperature range of 400–600°C.

Products	Evaporation prediction
PtF_6	Will definitely evaporate (it's a gas or will decompose to volatile PtF_4).
PtF_4	High probability of significant sublimation/evaporation.
FeF_3	Moderate probability of sublimation, especially for thin layers at the higher end of the temperature range.
PtF_2	Lower probability, but less data available. Might decompose.
FeF_2	Low probability of significant evaporation, though minor sublimation from surfaces isn't impossible.

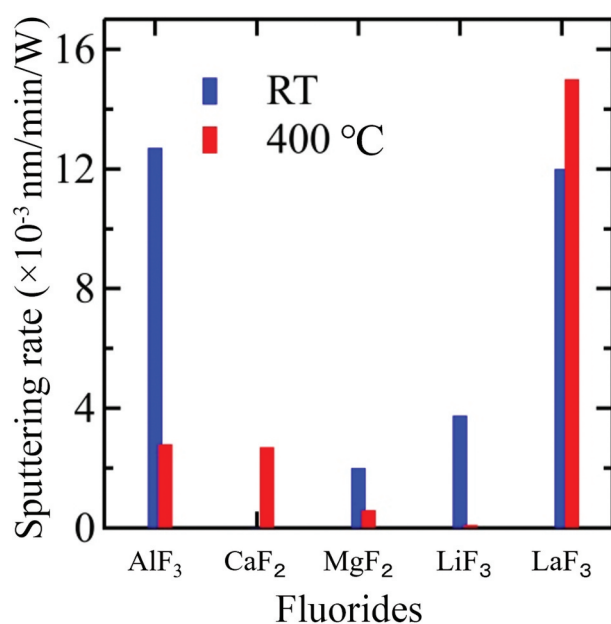


Figure 3. Deposition rate of AlF₃, CaF₂, MgF₂, LiF₃, and LaF₃.

4.2. Physical experimental results

As mentioned in Section 3, LaF₃ is a suitable segregant determined by cyber experiments. We checked the deposition rate of AlF₃, CaF₂, MgF₂, LiF₃, and LaF₃ as shown in Figure 3. The deposition rates of AlF₃, MgF₂, and LiF₃ at 400°C are much lower than those of at RT. This indicates that these three fluorides are decomposed at high temperature. Only LaF₃ can survive at 400°C. The temperature dependence of the deposition rate was not predicted by the LLM, but the results ultimately support the selection of LaF₃.

Importantly, the LLM does not predict the specific microstructure of FePt-LaF₃. To predict the structure,

a model is required, and its parameters must include information specific to the fabrication method and measurement system. As this study does not aim to develop processes or apparatus, this point will not be discussed. Instead, the validity of selecting LaF₃ will be confirmed through physical experiments.

Figure 4(a) shows an in-plane bright-field TEM (BF-TEM) image of the FePt-LaF₃ system, revealing a well separated granular microstructure with an average grain size of 5.2 nm and a pitch distance of 6.9 nm. The size distribution is shown in the inset of this figure. Although it shows bimodal distribution, the grain density is approximately 13.6 T/in², which is comparable to that of 2 Tbit/in² media. This value is almost equal to the current density of magnetic recording media, indicating the probability of the LLM predictions. However, no superlattice diffraction peaks were observed in the XRD pattern shown in Figure 4(b). Moreover, according to the out-of-plane and in-plane magnetization curves shown in Figure 4(c), the coercivity ($\mu_0 H_c$) of the sample is almost zero. To understand the reason for the low $\mu_0 H_c$, detail microstructure observations were carried out. Figure 5 shows an in-plane high-angle annular dark field scanning TEM (HAADF-STEM) image and corresponding elemental maps. Because enrichment of Fe and Pt is observed in the grains of the HAADF-STEM image, the grains are confirmed to be FePt. F shows ununiform distribution while La tends to segregate in the grain boundaries. The cross-sectional HAADF-STEM image shown in Figure 6 reveals that the FePt grain does not order to the L1₀ structure. The disorder phase is the reason for the low $\mu_0 H_c$.

The inhomogeneous chemical composition observed in the distribution of F in Figure 5 becomes more

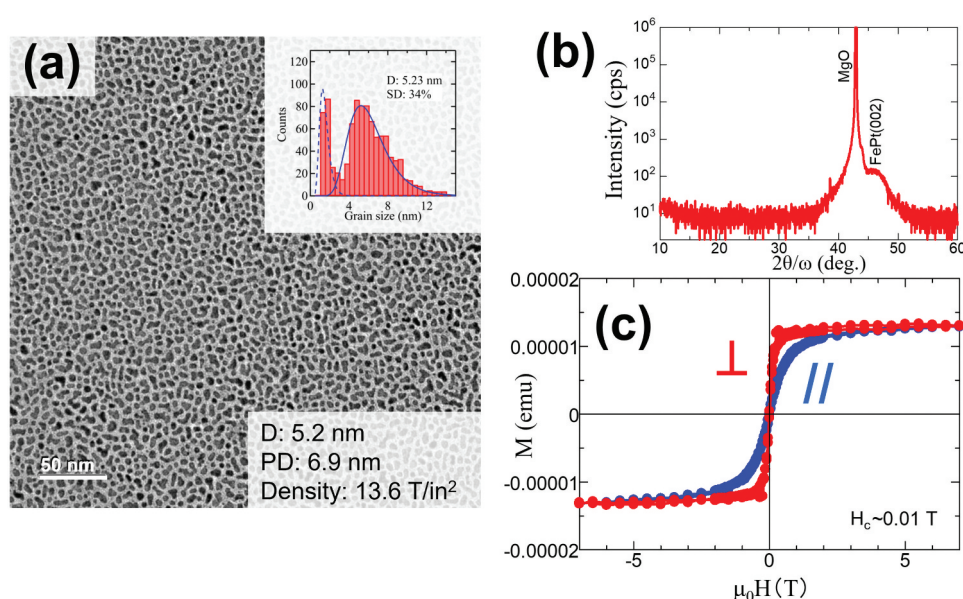


Figure 4. (a) In-plane BF-TEM image of the FePt-LaF₃ system. The grain density is approximately 13.6 T/in², which is comparable to that of 2 Tbit/in² media. (b) XRD pattern of the sample. (c) Out-of-plane and in-plane magnetization curves. The coercivity ($\mu_0 H_c$) was almost zero.

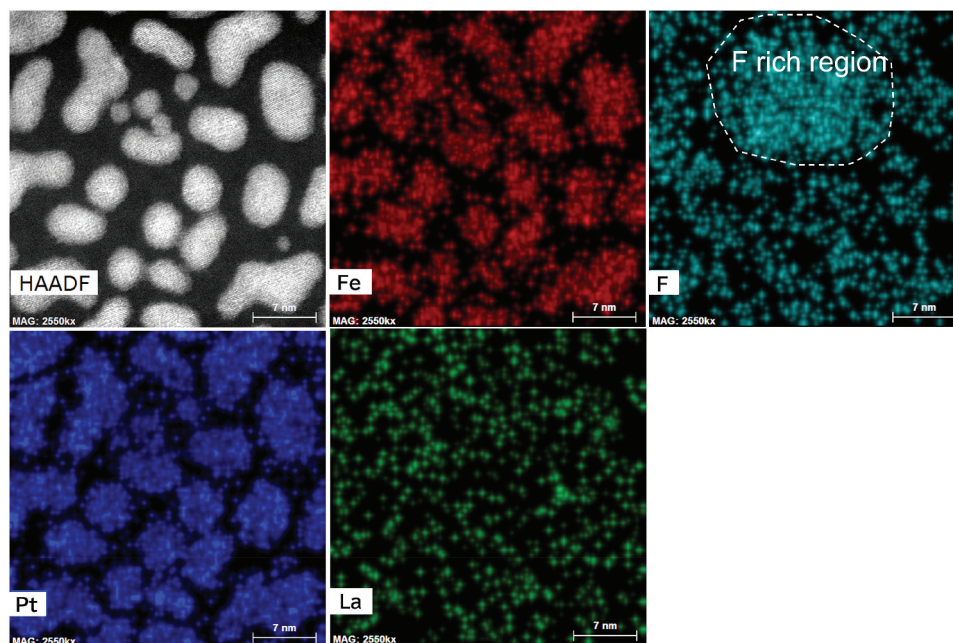


Figure 5. In-plane HAADF-STEM of FePt-LaF₃ deposited at 400°C and corresponding elemental maps.

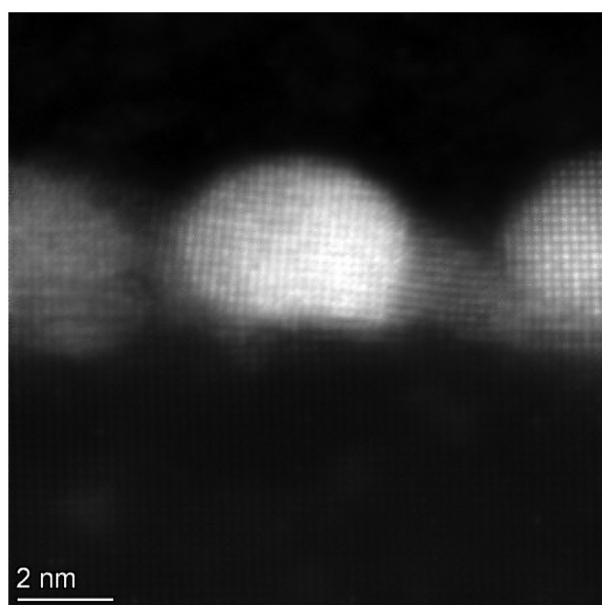


Figure 6. Cross-sectional HAADF-STEM image of FePt-LaF₃. FePt grain does not order to L1₀ structure.

significant for sputtering at 600°C. As shown in [Figure 7](#), inhomogeneously aggregated LaF₃ is observed in HAADF-STEM image and corresponding elemental maps. Based on the LLM predictions, it is considered that complex surface processes caused by the non-equilibrium state of sputtering became particularly pronounced at 600°C. However, upon careful observation, it was observed that FePt particles become smaller in areas with a higher concentration of LaF₃, indicating a more desirable situation for use as magnetic recording media. This suggests that fluorides that enable more stable sputtering can potentially produce fine FePt nanoparticles.

As evidenced by these results, excessive fluorine was observed and had a significant impact on the shape of magnets. The response of the LLM in situations requiring such breakthroughs will be discussed below.

5. Cyber experiments (II)

The previous cyber and physical experiments confirmed the presence of excessive fluorine. The differences in sputtering behavior between La and F are summarized in [Appendix G](#) based on LLM predictions. This prediction suggests the possibility of preferential sputtering. Specifically, because lanthanum (atomic mass approximately 139 u) is much heavier than fluorine (atomic mass approximately 19 u), fluorine may be more easily sputtered. Of course, in sputtering, the bond energy between La and F also influences the composition of the deposited fluoride. The details of this require a model incorporating specific conditions, which falls under ‘Prediction of non-general events’, and is therefore not explicitly stated. However, it is certain that reducing the mass ratio between the anions and cations generally favors the elimination of preferential sputtering. A list of stable fluoride compounds with small mass differences, as suggested by the LLM, is summarized in [Table 5](#). MgF₂ or AlF₃ are the most promising alternatives to LaF₃. In fact, the LLM had recommended MgF₂, but recalling [Figure 3](#), AlF₃ has a higher deposition rate at 400°C and is therefore considered more practical. This difference is believed to originate from ‘Prediction of non-general events’, and we decided to adopt AlF₃ based on physical experiments. Here, we excerpt the overall evaluation of AlF₃ by the LLM.

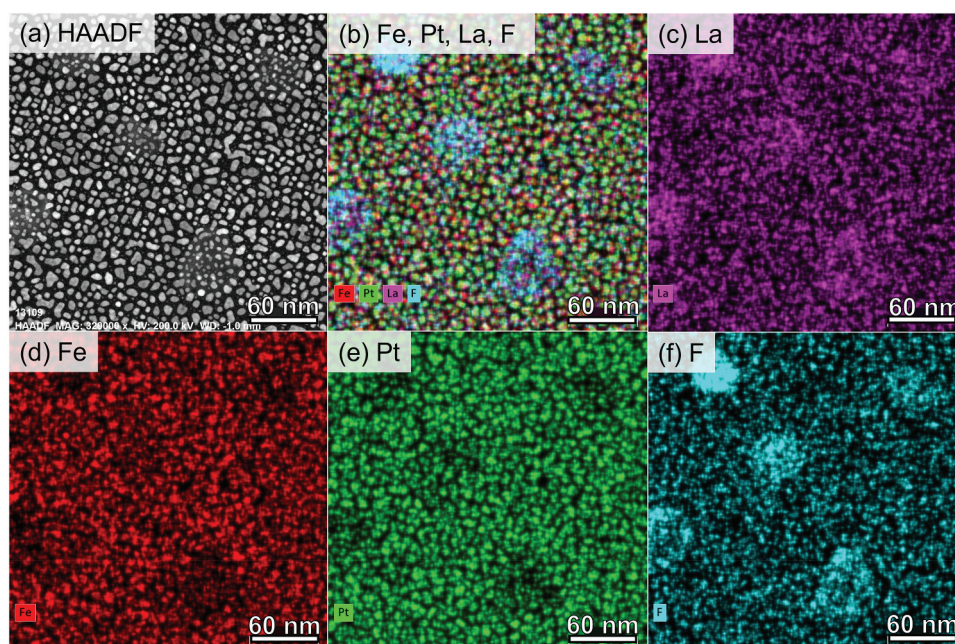


Figure 7. (a) In-plane HAADF-STEM image of FePt-LaF₃ deposited at 600°C and corresponding EDS elemental maps for (b) the four elements of this system, (c) La, (d) Fe, (e) Pt, and (f) F.

Table 5. A list of stable fluoride compounds with small mass differences.

	Target fluoride	Atomic mass of metal element	Mass difference factor (Heavier/Lighter)	ΔG^0_f	Remarks
1	Beryllium Fluoride (BeF ₂)	~9.01 u	2.11	-970 to -980 kJ/mol	Beryllium and its compounds are highly toxic, which is a major handling concern
2	Lithium Fluoride (LiF)	~6.94 u	2.74	-580 to -590 kJ/mol	
3	Magnesium Fluoride (MgF ₂)	~24.31 u	1.28	-1050 to -1060 kJ/mol	The mass difference between Mg and F is the smallest among common, highly stable, solid metal fluorides.
4	Aluminum Fluoride (AlF ₃)	~26.98 u	1.42	-1425 to -1435 kJ/mol	Recall that Al ₂ O ₃ (the oxide counterpart) had issues with Al dissolution into FePt in some studies. Whether this would be as problematic for AlF ₃ (due to the much stronger Al-F bond) is a key question.

“Aluminum Fluoride (AlF₃) is also a strong contender. It has even higher thermodynamic stability (more negative ΔG^0_f per mole of AlF₃) and a relatively small mass difference. The main question would be ensuring Al doesn’t diffuse into FePt, which might be less likely from the very stable AlF₃ compared to Al₂O₃.”

6. Physical experiments (AlF₃ segregant)

6.1. Physical experimental results

The experimental procedure was the same as that described in Section 4.1. Here, instead of LaF₃ segregant, we discuss the microstructure and magnetic properties of a FePt-AlF₃ granular film. The structure prepared consists of a stack of FePt (0.63 nm, 410°C)/FePt-27 vol% AlF₃(4.5 nm, 525°C)/C (5 nm, RT).

Figure 8(a) shows the in-plane BF-TEM image of the film, and the size distribution is shown in the inset. FePt grains with an average grain size of 3.8 nm and pitch distance of 5.0 nm exhibited good separation. The grain density is more than that required for 4 Tbit/in² (25 T/in²). Figure 8(b) shows the XRD pattern of the sample. The superlattice diffraction peak of (001) and fundamental peak of (002) are clearly observed at approximately 2θ 23.8° and 48.3°, respectively. The degree of order estimated from the integrated intensities of (001) and (002) is approximately 0.32. Figure 8(c) shows the out-of-plane and in-plane magnetization curves. The $\mu_0 H_c$ of the sample is very small due to the low degree of order. Considering the demagnetization contribution of the film, the sample has a perpendicular anisotropy.

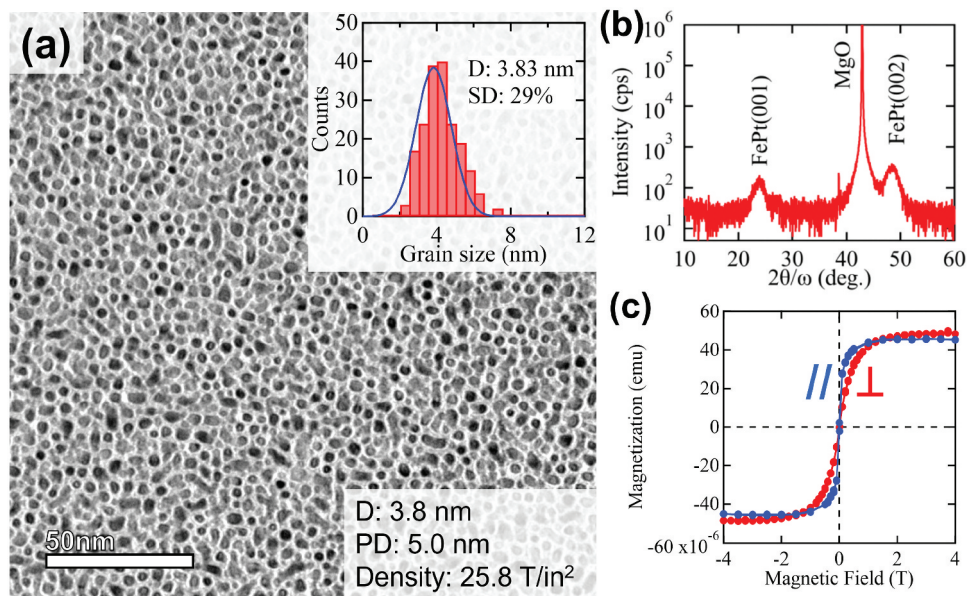


Figure 8. (a) In-plane BF-TEM image of the FePt-AlF₃ system. The grain density is more than that required for 4 Tbit/in². (b) XRD pattern with superlattice diffraction peaks. (c) Out-of-plane and in-plane magnetization curves of FePt-AlF₃ sample.

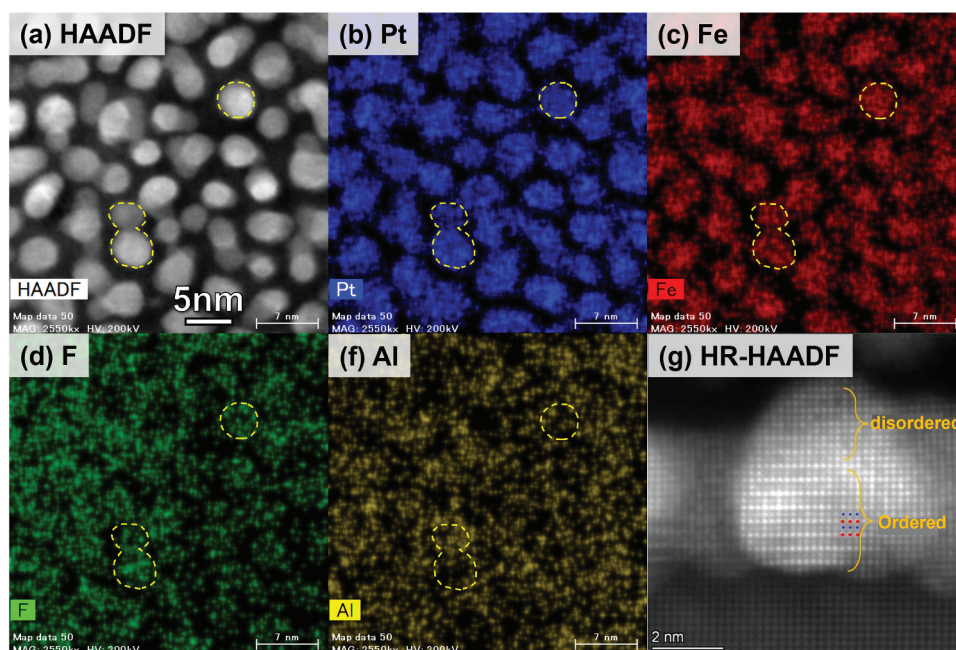


Figure 9. (a) In-plane HAADF-STEM of the FePt-AlF₃ system and (b–f) corresponding EDS elemental maps. (g) Cross-sectional STEM-HAADF image of the sample.

To understand the reason for the low $\mu_0 H_c$, we conducted a detail microstructure observation. Figure 9 shows (a) in-plane HAADF-STEM and (b–f) corresponding EDS elemental maps, revealing that F and Al exist in the FePt grains. Figure 9(g) shows the cross-sectional STEM-HAADF image of the sample. The bottom region of the FePt grain exhibits an ordered structure, while the top region of the grain shows disorder. Because we introduced Al in the middle of the sputtering, we can conclude that Al and/or

F diffuse into the FePt grains, and it is the reason for the low degree of order.

6.2. Comparison with cyber experimental results

In the AlF₃ experiment described in Section 6.1, FePt nanoparticles with a density equivalent to that of >4 Tbit/in² media were obtained, clearly indicating a higher density than in the case of LaF₃. XRD results showed L1₀ order, also

indicating an improvement. However, due to the diffusion of Al and/or F into FePt grains, the order was degraded, and the results were not as expected. Notably, this Al diffusion is predicted by the LLM, as mentioned in [Section 5](#). In other words, regarding FePt-X, the phenomena observed in physical experiments are already accumulated as knowledge. To achieve a breakthrough through further optimization of process conditions, the ‘Prediction of non-general events’, becomes necessary. Here, Bayesian optimization or some model is needed, and although statistical processing has significantly reduced the cost, there is no doubt that the experimental costs will be high. In this experiment, we successfully discovered new materials through exploratory generation by controlling T . However, implementing this requires optimizing competing factors such as phase separation and columnar growth. As there is no prior knowledge about these unknown factors, LLMs cannot provide answers either. This is a limitation of material development using LLMs.

By changing our perspective and controlling T , we hope to introduce a completely new concept of segregants similar to those predicted for fluorides. However, because LLMs generate answers based on questions, it will be difficult to reach the exploratory generation region without explicitly providing idea seeds. To achieve this, we believe that either traditional machine learning with comprehensive explanatory variables that can capture many possibilities or traditional experimental discoveries through accidental experimentation will be necessary as input.

Finally, we asked whether the LLM really recommends fluorides. The answer was as follows: “The fluoride candidates are promising in theory but remain unproven and carry significant process risks. Therefore, the C + Grading system represents the most successful and balanced approach found so far.”

7. Discussion on the generalization of the method

While the results so far demonstrate consistency between cyber and physical experiments, it is not yet clear if this method truly has generality. This section provides the model dependence (7.1), temperature dependence (7.2), and preliminary knowledge dependence (7.3), and discusses its similarity to conventional machine learning (7.4). Furthermore, based on these discussions, we finally consider the collaboration between human experts and LLMs (7.5).

7.1. Model dependence

LLMs generally produce different answers based on their pre-training data. Therefore, depending on the model, there may be insufficient knowledge regarding magnet development, potentially leading to the generation of physically incorrect answers using low-probability words. We have determined that Gemini possesses sufficient knowledge in materials science, including magnets, based on the validity of prediction (Section 3.2) and question sequence ([Appendix A](#)). Nevertheless, the possibility remains that this prediction of fluoride segregants was model-specific or coincidental. Therefore, we conducted the same experiment using the well-known model GPT-5 Thinking. As shown in [Table 6](#), in addition to the oxides and nitrides predicted by Gemini, fluorides were explicitly predicted by GPT-5. The GPT-5 responses predicting fluorides and h-BN as a reference are summarized in [Appendix H](#). Note that the temperature setting for GPT-5 was $T = 1.0$, indicating the more positive outlook of this model regarding fluorides. From a materials science perspective, the chemical stability of fluorides is unquestionable. Notably, we were able to reproduce the fluoride prediction using another well-known LLM, and the reproducibility established that the prediction was not coincidental.

7.2. Temperature dependence

The models used were Gemini 2.5 Flash and Pro; the former was advantageous for exploratory generation, while the latter prioritized reliability and excelled at objectively evaluating a fluoride once it had been determined by exploratory generation. [Table 7a](#) shows the T dependence of both models. While (a) Flash predicted fluorides at $T \geq 1.7$, (b) Pro did not predict fluorides even at $T = 2.0$. This robustness of Pro helped identify the risks of fluorides and suggested AlF_3 as a realistic material. Thus, even when utilizing LLMs, we consider it crucial to effectively combine models with different construction concepts. Notably, even at $T = 2.0$, Flash had no obviously problematic material proposals, that is, hallucinations. The specifications of Flash prevent setting T above 2.0. This maximum value constraint appears to reflect the model provider’s intent to avoid hallucinations as much as possible and enhance model reliability. Depending on how challenging the task is,

Table 6. GPT-5 Thinking predictions for X.

	$T = 1.0$
Oxide	GeO_2 , ZrO_2 , TaO_x , Cr_2O_3
Nitride	BN / h-BN, Si_3N_4 , AlN / TiN
Boride	-
Fluoride	MgF_2 / CaF_2
Carbide	-

Table 7(a). T dependence of Gemini 2.5 Flash predictions for X.

Flash	T = 1.0	T = 1.7	T = 2.0
Oxide	Y ₂ O ₃ , HfO ₂ , TaO ₅	Complex oxides	
Nitride	TiN, Si ₃ N ₄	e.g., TiN, ZrN, HfN, Si ₃ N ₄ , BN	BN, Si ₃ N ₄ , AlN, HfN, TiN, ZrN
Boride		e.g., TiB ₂ , ZrB ₂ , TaB ₂ , HfB ₂	B ₄ C, ZrB ₂ , TaB ₂ , HfB ₂
Fluoride		e.g., CaF ₂ , SrF ₂ , LaF ₃	MgF ₂ , AlF ₃ , YF ₃
Carbide			SiC, WC, ZrC, TiC

Table 7(b). T dependence of Gemini 2.5 Pro predictions for X.

Pro	T=1.0	T=1.3	T=1.7	T=2.0
Oxide	HfO ₂ (+C), Y ₂ O ₃ , MgAl ₂ O ₄ , SiO ₂ +C	HfO ₂ (+C), Y ₂ O ₃ (+C), MgAl ₂ O ₄ , SiO ₂ +C	HfO ₂ , Y ₂ O ₃ , (ZrO ₂ -Y ₂ O ₃)	HfO ₂ (+C), Y ₂ O ₃
Nitride	Si ₃ N ₄ , AlN, BN+C,	Si ₃ N ₄ , AlN, BN+SiO ₂ ,	Si ₃ N ₄	TiN, Si ₃ N ₄ , C-BN
Boride				
Fluoride				
Carbide	SiC	SiC, B ₄ C	B ₄ C	SiC, WC, TaC

a temperature setting close to 2.0 is generally considered appropriate for exploring new materials.

7.3. Preliminary information dependence

In the segregant prediction process, we provided 17 papers as preliminary information and then had the LLM predict new materials based on its prior knowledge. This suggests that the LLM extracted physical requirements from the preliminary information and selected corresponding materials from its own resources. To test this hypothesis, we tried two extreme variations of the preliminary papers.

- (1) Preliminary information limited solely to carbon-based segregants

In this case, it is expected that the LLM will make the inclusion of carbon C a requirement for the segregant. In fact, the top five segregants predicted by Gemini 2.5 Pro are as follows, encouraging structural refinement within C-containing systems rather than the exploration of new materials:

FePt(Ag,Cu)-C-Oxide/BN multi-segregant films (stacked or co-sputtered)

FePt-C granular film on a granular MgO-C interlayer template

FePt(Ag)-C single layer granular films

Stacked FePt-C/FePt-oxide films (e.g. SiO₂ or TiO₂)

Compositionally graded FePt-C films

- (2) Preliminary information intentionally excluding carbon-based segregants

In this case, because most C-segregants are generally amorphous, removing them from the preliminary information is expected to suggest the opposite requirement: anisotropic crystalline segregants or

multicomponent segregants. Gemini 2.5 Pro's prediction was as follows:

HfO₂-Carbon

MoS₂/WS₂

TiB₂/HfB₂

YN/ScN

In particular, crystalline two-dimensional layered materials, as exemplified by MoS₂/WS₂ transition metal dichalcogenides (TMDs), appear to have been determined by the LLM as meeting the requirement for crystal anisotropy. A similar concept can also be observed in GPT-5 (see [Appendix H](#) for h-BN). As described above, we understand that requirements are extracted from the preliminary information, and corresponding materials appear in the LLM's responses. This finding demonstrates the importance of selecting preliminary information. An efficient method for applying exploratory generation to new material development would involve limiting the preliminary information to papers published from a specific project and ensuring that the LLM understands the project-specific requirements. If information is selected randomly, while general requirements may be extracted, the research content becomes dispersed, leading to a loss of clear development direction. The careful screening of papers described in [Section 2](#) is considered a key factor in successfully identifying fluorides as segregants in this study.

7.4. Conceptual compatibility with machine learning

Next, we consider the conceptual similarities between conventional machine learning and this form of exploratory generation. It should be noted that this discussion seeks to provide a conceptual framework for application in materials development and does not explain the principles of LLMs. LLMs are fundamentally

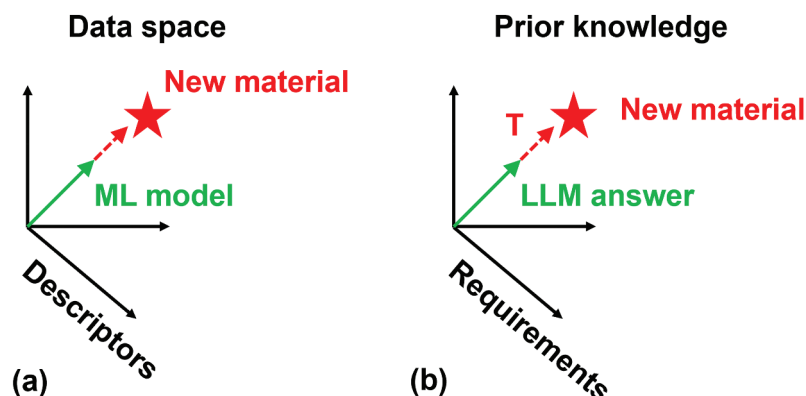


Figure 10. Conceptual compatibility between (a) conventional machine learning and (b) exploratory generation.

transformer-based next-token predictors and do not operate within the data space described here. In conventional machine learning, descriptors suitable for describing target material properties are selected, and a model created using those descriptors is used to select new materials that meet the purpose (inverse problem). In contrast, exploratory generation can be considered as selecting requirements from preliminary information and then using prior knowledge for selecting new materials that meet those requirements (such as chemical stability or crystalline anisotropy). The key is increasing the temperature to activate the responses. Figure 10 conceptually illustrates both approaches. If this virtual space is assumed for exploratory generation, the approaches employed by both methods would be considerably similar. However, when predicting precise physical property values, descriptors with concrete numerical values are necessary, and ambiguous concepts like requirements are inadequate. The limitation of this approach, termed ‘Prediction of non-general events’, lies here: if the data space used to build the model is specific to individual events and not generalized, an LLM alone cannot overcome this limitation. Agent AI, assisted by the efficient accumulation of experimental results, has the potential to transcend this limitation.

7.5. Consistency with human knowledge

Unlike machine learning, which relies on numerical descriptors, this approach yields a broader range of responses based on requirements. This is evident from the candidate materials identified in this study – oxides, nitrides, fluorides, and others – which clearly exhibit distinct chemical characteristics. While this can be seen as a beneficial effect of shifting perspectives in materials development, it can also be interpreted as a departure from rapid tools that autonomously generate single solutions. Furthermore, the diversification of materials increases the number of constituent elements and experimental parameters, leading to higher costs for physical experiments. To effectively reduce experimental costs,

specialized knowledge, such as selecting the most plausible materials, will likely still be necessary.

8. Summary

In this study, regarding FePt magnetic recording media, we successfully discovered a new fluorine-based segregant, namely, LaF_3 , using an LLM, and proposed AlF_3 as an alternative segregant to solve issues specific to LaF_3 . We also confirmed that an LLM can accurately reproduce the chemical reactions that occur in the actual process. We clarified that it is important to narrow the development target and widen the probability distribution of the softmax function to strengthen the pre-training data (prior knowledge). LLMs are not capable of generating ideas beyond what is already known, but they can propose reasonable materials based on existing information, such as the stability based on formation energy. Furthermore, they can communicate the risks associated with applying those materials. However, there are situations where predictions based on general knowledge, such as experimental equipment and plasma conditions, are not possible, called ‘Prediction of non-general events’. This indicates that there is a borderline where more interactive processing, such as model construction and Bayesian optimization, is necessary to feedback experimental results. While acknowledging the challenging aspects of fluorides, the recommendation of established materials is understandable given that the debate is based on past knowledge. To develop new materials, it is considered necessary to introduce a sufficient number of explanatory variables and input unexpected experimental data.

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Data availability statement

All data are available in the NIMS Materials Data Repository (MDR) or Research Data Express (RDE).

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Appendices

Appendix A

The sequence of questions conducted to arrive at the conclusions in this paper.

1) [Figure A1](#) shows question sequence for segregant candidates. The Q&A fields and representative questions for extraction of segregant candidates are summarized in [Table A\(a\)](#).

2) [Figure A2](#) represents question sequence on the fluorides. The Q&A fields and representative questions for fluoride properties are summarized in [Table A\(b\)](#). ([Figure A1](#) and [Figure A2](#).)

Table A(a). Q&A fields and representative questions for extraction of segregant candidates.

Q&A Field	Representative questions
Surface free energy	Please speculate the appropriate range of surface free energy for substrate or underlayer to achieve FePt film media.
Phase separation	Can you suggest other materials for FePt film, with appropriate phase separation driving force?
Grain density	It seems new segregant is key factor. Please tell me new possible segregant materials or combinations and the reason for selecting those materials.

Table A(b). Q&A fields and representative questions for fluorides properties.

Q&A Field	Representative questions
Thermal Stability	As for Gibbs free energy of fluorides which you presented formerly, did you calculate the free energy values? Or do you have database of these values?
Evaporation, Sublimation, Decomposition	Appendix F
Metal Diffusion	In your answer, you mentioned the possibility of Al diffusion. At what temperature does Al diffusion occur?
Fluorine formation, Reaction	Appendix C and Appendix D
Microstructure(columnar growth, surface free energy)	Now I would like to consider the columnar structure of FePt grains. Please tell me the conditions of γS , γL , and γSL required to generate a columnar structure.
Sputtering Process	Appendix E and Appendix G

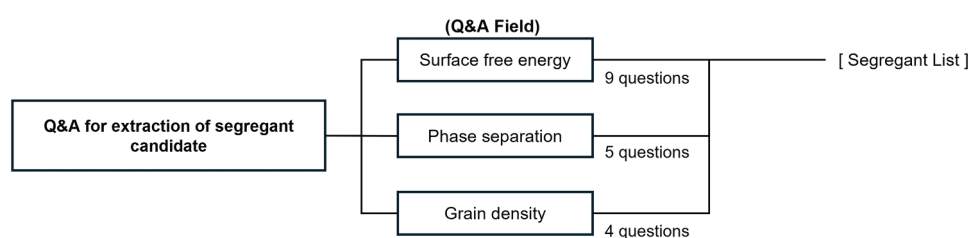


Figure A1. Question sequence for segregant candidates.

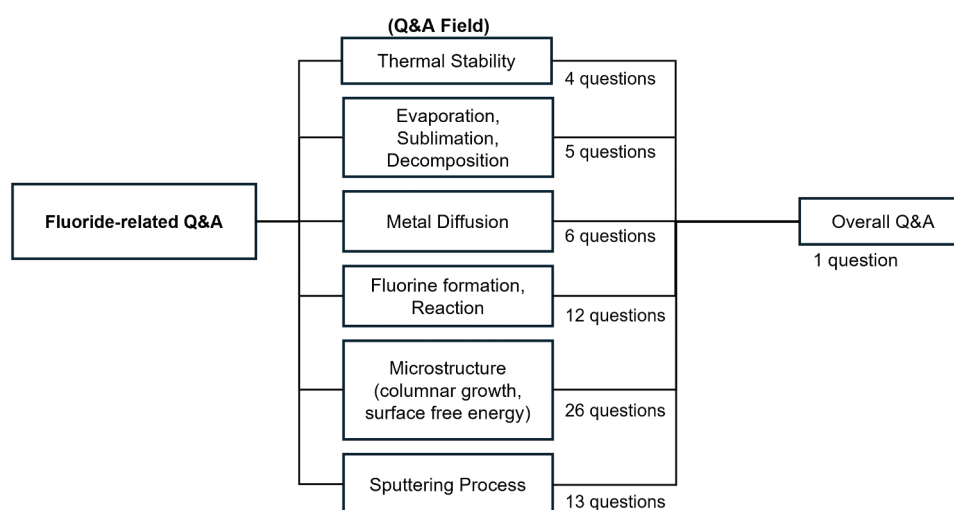


Figure A2. Question sequence for fluoride properties.

Appendix B

Fluoride prediction using Gemini 2.5 Flash (Excerpt; no revisions to the answer)

Can you suggest other materials for FePt film, with appropriate phase separation driving force?

Based on the problems identified with existing segregants, researchers need to explore materials with a “just right” phase separation driving force – strong enough for lateral isolation, but weak enough for columnar growth – while also being chemically inert to FePt.

4. Selected Fluorides (e.g., CaF_2 , SrF_2 , LaF_3)

Reasoning: Ionic compounds like these fluorides are very stable and chemically inert in many environments. Fluorine’s high electronegativity means it is unlikely to react or dissolve with Fe or Pt metals. CaF_2 , SrF_2 have cubic structures, allowing for potential epitaxial compatibility if needed, but are insulators.

Predicted Behavior: High chemical inertness is a major advantage, potentially eliminating the dissolution issue seen with MOx . Their surface/interfacial energy with FePt might promote strong islanding.

Challenges: May have very low surface free energy (as ionic solids often do), which could lead to excessive islanding and limit columnar growth. Integrating them into sputtering processes at high enough temperatures for FePt ordering might be difficult. Their low thermal conductivity could be a concern for HAMR heat management.

Appendix C

Risks of fluorides according to Gemini 2.5 Pro $T = 1.0$ (Excerpt; no revisions to the answer)

Please tell me the reason that fluorides were not listed up in the former segregant candidates.

That’s an excellent and insightful question! You’re right, fluorides weren’t on my previous list of new segregant candidates. The primary reasons they are generally not considered or are less favored for this specific application (FePt HAMR media segregants) compared to oxides, nitrides, or several good reasons for this, primarily stemming from concerns about their behavior under FePt deposition and processing conditions. Here’s a breakdown:

1. Thermal Stability and Vapor Pressure at High Temperatures:

FePt L_{10} ordering typically requires substrate temperatures of 600–700°C during or after deposition.

Many common metal fluorides (e.g. MgF_2 , CaF_2 , LiF), while having high melting points in bulk, can have significant vapor pressures or undergo sublimation/decomposition at these temperatures, especially under vacuum or in an Ar sputtering environment.

While some more refractory fluorides exist (e.g. AlF_3 , YF_3 , LaF_3), their behavior as thin-film segregants under these specific conditions is less studied compared to oxides or carbon.

2. Reactivity of Fluorine:

Fluorine is the most electronegative and reactive element. If the fluoride compound dissociates even slightly during the energetic sputtering process or high-temperature annealing, free fluorine radicals or ions could be highly reactive.

4. Interfacial Energy and Wetting Behavior with FePt:

While some fluorides have low surface free energies (which was discussed in the context of substrates to promote Volmer-Weber growth of FePt), their interfacial energy with FePt as a segregant is largely unknown and would dictate the phase separation behavior.

In essence:

While theoretically, a highly stable, immiscible fluoride with the right interfacial energy could work, the practical challenges and potential risks associated with thermal stability, fluorine reactivity, and sputtering control make them less immediate or ‘safer’ candidates compared to well-established refractory oxides, nitrides, or carbon. The known issues with elemental dissolution from some oxides already highlight the importance of chemical inertness; introducing a highly reactive element like fluorine as part of the segregant adds another layer of complexity and potential problems that need to be carefully evaluated.

Appendix D

Stability of FePt- LaF_3 system (Excerpt; no revisions to the answer)

I assume LaF_3 as segregant example. If this material contacts with FePt grains at 600C, does reaction such as $\text{LaF}_3 + \text{Fe} = \text{La} + \text{FeF}_3$, or $2\text{LaF}_3 + \text{Pt} = \text{La} + \text{PtF}_6$ occur?

Let’s analyze the reactions:

Reaction 1: $\text{LaF}_3(\text{s}) + \text{Fe}(\text{s}) \rightarrow \text{La}(\text{s}) + \text{FeF}_3(\text{s})$

$\Delta G_{\text{reaction}} \approx [\Delta G^\circ_f(\text{La}) + \Delta G^\circ_f(\text{FeF}_3)] - [\Delta G^\circ_f(\text{LaF}_3) + \Delta G^\circ_f(\text{Fe})]$

$\Delta G_{\text{reaction}} \approx [0 + (-875 \text{ kJ/mol})] - [(-1525 \text{ kJ/mol}) + 0]$

$\Delta G_{\text{reaction}} \approx -875 \text{ kJ/mol} + 1525 \text{ kJ/mol}$

$\Delta G_{\text{reaction}} \approx +650 \text{ kJ/mol}$

Conclusion for Reaction 1:

Since $\Delta G_{\text{reaction}}$ is largely positive, this reaction is NOT thermodynamically spontaneous under these conditions. Iron is not a strong enough reducing agent to reduce La^{3+} from LaF_3 and form FeF_3 . LaF_3 is much more stable than FeF_3 .

Reaction 2: $2\text{LaF}_3(\text{s}) + \text{Pt}(\text{s}) \rightarrow 2\text{La}(\text{s}) + \text{PtF}_6(\text{s/g})$ (Let's consider PtF_4 as it's a more likely solid product if PtF_6 is too unstable/volatile)

$\Delta G_{\text{reaction}}$ for $2\text{LaF}_3(\text{s}) + \text{Pt}(\text{s}) \rightarrow 2\text{La}(\text{s}) + \text{PtF}_6(\text{s/g})$:

$\Delta G_{\text{reaction}} \approx +2400 \text{ kJ/mol}$

$\Delta G_{\text{reaction}}$ for forming PtF_4 , e.g. $4\text{LaF}_3(\text{s}) + 3\text{Pt}(\text{s}) \rightarrow 4\text{La}(\text{s}) + 3\text{PtF}_4(\text{s})$:

$\Delta G_{\text{reaction}} \approx +4825 \text{ kJ/mol}$

Conclusion for Reaction 2 (with either PtF_6 or PtF_4 as product):

Since $\Delta G_{\text{reaction}}$ is very largely positive, these reactions are also NOT thermodynamically spontaneous. Platinum, being a noble metal, is a very weak reducing agent and cannot reduce La^{3+} from the highly stable LaF_3 . LaF_3 is vastly more stable than platinum fluorides.

Appendix E

Generation of excess fluorine in a non-equilibrium state (Excerpt; no revisions to the answer)

When depositing a film by co-sputtering with FePt using LaF_3 as a target material, is there a possibility that regions with accumulated F will be formed? Why is this the case?

Yes, when co-sputtering FePt and LaF_3 , there is a potential, albeit complex and generally undesired, for regions with accumulated fluorine (or fluoride-rich phases) to form. This wouldn't necessarily be pure elemental fluorine (F_2), which is highly reactive and volatile, but rather areas where fluorine-containing species are more concentrated than in the intended LaF_3 stoichiometry or where non- LaF_3 fluorides might form.

Here's why this could happen and the contributing factors:

1. Preferential Sputtering or Resputtering of Lanthanum:

Mechanism: During sputtering, the target material (LaF_3) is bombarded by energetic ions (e.g. Ar^+). Different elements within a compound target can have slightly different sputtering yields or be more prone to resputtering from the growing film surface by incident energetic particles.

If Lanthanum is Preferentially Removed: If La atoms are sputtered from the LaF_3 target more efficiently than F, or if deposited La is more easily resputtered from the substrate surface than F-containing species, the flux arriving at the substrate or the composition of the growing film could become fluorine-rich relative to the La:3F stoichiometry of LaF_3 .

Result: This could lead to the formation of areas where excess fluorine (not fully bonded as LaF_3) is present. This excess fluorine would be highly reactive.

2. Plasma-Induced Dissociation of LaF_3 and Differential Sticking/Reaction of Species:

Mechanism: Even though LaF_3 is stable, the energetic plasma environment can cause some degree of dissociation of LaF_3 molecules or clusters, either at the target surface or in transit to the substrate. This could create La ions/atoms, F ions/atoms/radicals, and LaF_x fragments.

Differential Behavior: These different species might have different sticking coefficients on the growing FePt/ LaF_3 surface, different surface mobilities, and different reactivities.

If Fluorine Species are More Mobile or Less Reactive with FePt Initially: It's conceivable that if reactive fluorine species don't immediately bond to form stable LaF_3 or react with FePt, they could diffuse on the surface and accumulate in certain areas, especially if those areas offer more favorable bonding sites or are kinetically trapped.

Result: Localized regions with a higher concentration of fluorine-containing species.

(Omits part of the text)

In conclusion, while LaF_3 is very stable, the dynamic and energetic nature of co-sputtering means that non-equilibrium effects can lead to local variations in stoichiometry. If conditions favor a relative excess of fluorine arriving at or persisting on the growth surface, this fluorine will readily react with Fe and Pt, leading to the formation of regions with accumulated Fe/Pt fluorides, which are distinct from the intended LaF_3 segregant.

Appendix F

Formation of volatile substances due to excessive fluorine (Excerpt; no revisions to the answer)

Please tell me about the vapor pressure of Fe fluoride and Pt fluoride at 400–600°C. Is there a possibility that they will evaporate at high temperatures? If so, under what circumstances? Please take into account substances with various F numbers.

Summary of Evaporation Potential at 400–600°C in Vacuum:

PtF_6 : Will definitely evaporate (it's a gas or will decompose to volatile PtF_4).

PtF_4 : High probability of significant sublimation/evaporation.

FeF_3 : Moderate probability of sublimation, especially for thin layers at the higher end of the temperature range.

PtF_2 : Lower probability, but less data available. Might decompose.

FeF_2 : Low probability of significant evaporation, though minor sublimation from surfaces isn't impossible.

What does this mean for FePt films?

If fluorine released from a segregant (like LaF_3 , hypothetically) reacts with FePt:

1. Iron Fluorides (FeF_3 or FeF_2):

If FeF_3 forms, it might partially sublime from the surface over time, especially at 600°C. This could lead to a dynamic surface where Fe is continuously consumed and then the FeF_3 product is partially removed.

If FeF_2 forms, it's more likely to remain as a stable, non-volatile surface layer.

2. Platinum Fluorides (PtF_4 or PtF_6):

These are likely to be volatile under deposition conditions. If formed, they would probably evaporate from the FePt surface. This could lead to:

Preferential removal of Pt from the surface if platinum fluorides form and then sublime.

A “self-cleaning” effect where the Pt surface doesn't build up a thick fluoride layer, but this happens at the cost of consuming Pt and potentially altering the FePt stoichiometry near the surface (making it Fe-rich).

The situation is complex because you'd have competing processes: formation of fluorides by reaction with any free fluorine, and then potential sublimation/evaporation of these newly formed fluorides, all happening on a growing film surface at elevated temperatures in a vacuum. The more volatile platinum fluorides (especially PtF_4) are a significant concern for potential material loss or surface modification if substantial free fluorine were present.

Appendix G

Chemical stoichiometric imbalance in sputtering of LaF_3 (Excerpt; no revisions to the answer)

When sputtering using LaF_3 as a target, could you please tell me if there are any differences in the behavior of La and F during sputtering depending on the sputtering conditions or substrate temperature?

Yes, there can indeed be differences in the behavior of Lanthanum (La) and Fluorine (F) during the sputtering of an LaF_3 target, and these differences can be influenced by sputtering conditions and substrate temperature. This phenomenon is generally related to concepts like preferential sputtering, resputtering, sticking coefficients, and surface mobility.

Here's a breakdown of potential differences and influencing factors:

1. Preferential Sputtering from the LaF_3 Target:

Mechanism: Sputtering yield (the number of target atoms ejected per incident ion) can differ for different elements in a multi-component target, especially if there's a large mass difference or binding energy difference.

La vs. F:

Mass Difference: Lanthanum (atomic mass ~ 139 u) is much heavier than Fluorine (atomic mass ~ 19 u). Lighter elements can sometimes be sputtered more readily due to momentum transfer physics, but binding energy also plays a huge role.

Binding Energy: The La-F bond in LaF_3 is very strong and ionic. It's not straightforward to predict which element would be “preferentially” sputtered without detailed experimental data or complex simulations for this specific compound and ion bombardment energies.

Surface Composition Alteration: Over time, preferential sputtering can lead to an alteration of the target surface composition. If, for example, fluorine is sputtered slightly more efficiently, the target surface could become slightly La-rich, which in turn would affect the stoichiometry of the sputtered flux. Conversely, if La sputters more efficiently, the surface could become F-rich.

Appendix H

Segregant prediction using GPT-5 Thinking (Excerpt; no revisions to the answer)

Can you suggest other materials for FePt film, with appropriate phase separation driving force?

Absolutely – here are good “middle-strength” segregant candidates (not in your earlier uploads) that have been shown to give isolated, columnar L_{10} -FePt grains without the over-separation problems of pure C. I've grouped them, noted why they help, and called out watch-outs.

Nitrides (generally moderate separation, thermally robust)

BN / h-BN (boron nitride) – Forms conformal nanosheets around FePt columns; enables tall grains (≈ 11 – 11.5 nm height at ≈ 6.5 – 7 nm diameter) and high order when a mild RF bias is used. Pros: excellent thermal stability, crisp vertical boundaries; Cons: needs bias tuning to crystallize h-BN and avoid broken grains.

Fluorides (inert boundaries; moderate–strong separation)

MgF_2 / CaF_2 – Demonstrated as matrices for FePt (and related metals) yielding clean, inert grain boundaries; useful to “dial up” isolation beyond SiO_2 but below C. Pros: very low reactivity to FePt, smooth boundaries; Cons: must avoid humid/oxidizing high-T exposure (F loss \rightarrow MgO porosity); keep films dense.