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PERSPECTIVE PIECE

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Creating multifunctionality with hierarchical heterostructures

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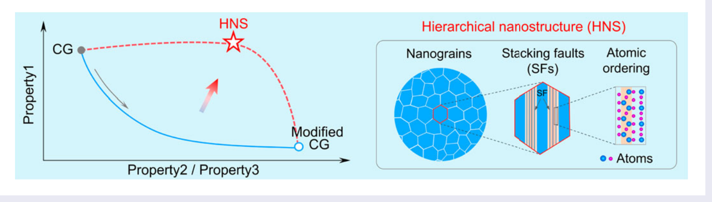
ABSTRACT
Materials with multifunctionalities are essential for saving energy and conserving natural resources, but intrinsic multiple trade-offs among material properties often limit the development of next-generation multifunctional materials. This Perspective highlights the concept of hierarchical heterostructure with the potential to create unprecedented multifunctional properties. This strategy enables an exceptional combination of multiple conflicting properties beyond the reach of conventional approaches through activating multiple physical mechanisms. This paper aims to draw the attention of materials community to the emerging heterostructured materials and to motivate active investigations on this topic for creating transformative materials with multifunctionalities.



IMPACT STATEMENT
Engineering hierarchical heterostructures opens up new opportunities to create multifunctionalities that are unattainable using conventional approaches.

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Background

Multifunctional materials are increasingly needed to meet the growing global demands for energy efficiency, resource conservation, and exceptional performances across a wide range of critical applications [1,2]. However, creating multifunctional materials presents a significant challenge due to the trade-offs among multiple material properties. Traditional alloying strategies often enhance one attribute at the expense of others [2,3]. For example, in the case of ferromagnetic materials that are critical for energy conversion and transmission [2,4], increasing electrical resistivity with non-magnetic components, necessary for avoiding heating by eddy currents [2], typically compromises saturation magnetization (M_s), thereby reducing energy density [5] (Figure 1). Similarly, enhancing coercivity and its thermal stability usually necessitate the inclusion of non- and weak-magnetic components or uses expensive heavy rare-earth elements to resist magnetization reversal [6], also reducing the M_s and thus energy density (Figure 1). As a result, achieving a multifunctional ferromagnetic material with high energy density, large electrical resistivity, and high thermal stability of coercivity remains elusive.

Heterostructuring is a new concept and strategy to address various tricky trade-offs in both structural and functional materials [7–9]. Despite great advances in the synthesis of heterostructured materials [7], however, most approaches reported so far typically focus on navigating only one trade-off between two material properties by constructing heterostructures. Simultaneously addressing multiple trade-offs in multiple properties remains a great challenge.

It is reasoned that constructing heterostructured materials with hierarchical structure could circumvent this dilemma [7]. This strategy makes it possible to synergistically control magnetic, electrical, thermal, and optical properties through the effective manipulation of domain wall movement, electron and phonon transport, as well as light propagation with abundant interfacial effects, leading to multifunctional properties.

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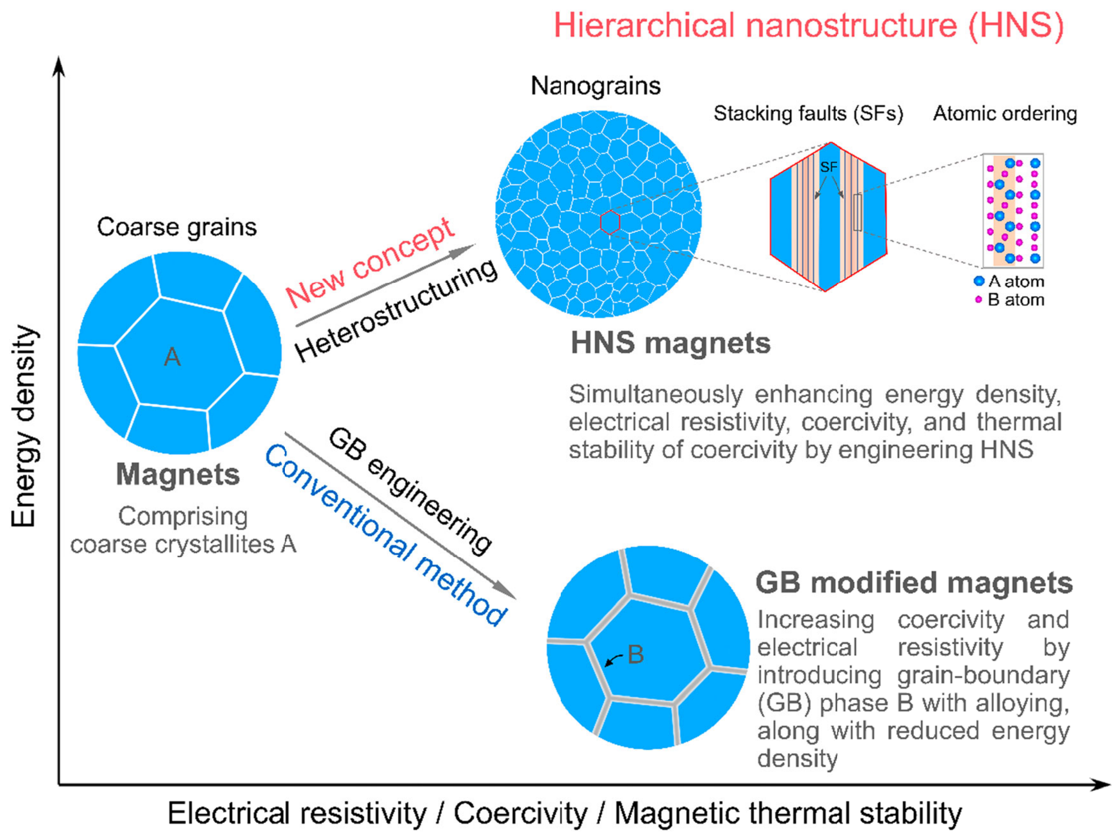

Figure 1. Hierarchical nanostructure (HNS, a type of hierarchical heterostructure) produces multifunctionalities. The electrical resistivity, coercive, and magnetic thermal stability of permanent-magnet materials are typically enhanced by introducing grain-boundary (GB) phase with alloying or adding non-magnetic components at the expense of energy density, owing to reduced magnetization. Constructing HNS enables a synergic enhancement of these conflicting properties through activating various physical mechanisms to simultaneously resist domain wall movement and electron transport, yielding the desired multifunctionalities.

A recent pioneering work demonstrated the success of this strategy of hierarchical heterostructure to create multifunctional materials, in which a significant breakthrough in multifunctional ferromagnets was achieved by engineering hierarchical nanostructures (HNSs) [10]. The HNSs embody a cascade of ordered structures, ranging from aligned nanograins on the tens-of-nanometers scale to nanoscale stacking faults within the grains and atomic-level elemental arrangements (see Figure 1). It opens up new opportunities to create multifunctional materials with unprecedented properties through constructing hierarchical heterostructures. This concept can be extended to other materials systems to realize multifunctionality, including thermoelectric, ferroelectric, and catalytic materials, where trade-offs among various properties are common.

New HNS ferromagnets with multifunctionalities

As an example of hierarchical heterostructured materials, the newly developed HNS magnets [10] comprise aligned $PrCo_5$ nano-grains (along the easy magnetization axis) embedded with high-density stacking faults (SFs), accompanying with atomic-scale local composition variations (see the schematic in Figure 1). The resulting HNS magnets exhibit a superior combination of high energy density, $(BH)_{max}$, and large coercivity (H_{ci}), far beyond that of previously reported $PrCo_5$ magnets (Figure 2(a)). They also have an exceptional combination of high $(BH)_{max}$ and high electrical resistivity (ρ) as compared with that of conventional $PrCo_5$ (CGS), commercial $SmCo_5$ and Sm_2Co_{17}-type high-temperature permanent magnets (Figure 2(b)). Moreover, the new HNS magnets demonstrate an unprecedentedly high thermal stability of coercivity (indicated by $-1/\beta$, where β is the temperature coefficient of H_{ci}), greatly surpassing that of existing commercial rare-earth magnets (Figure 2(c)). Consequently, despite of their mutually conflicting attributes, the HNS strategy enables a synergetic enhancement of the $(BH)_{max}$, H_{ci}, ρ, and $-1/\beta$ that is far beyond the reach of conventional methods. This synergic enhancement was realized through creating the distinctive hierarchical heterostructure that cascades from

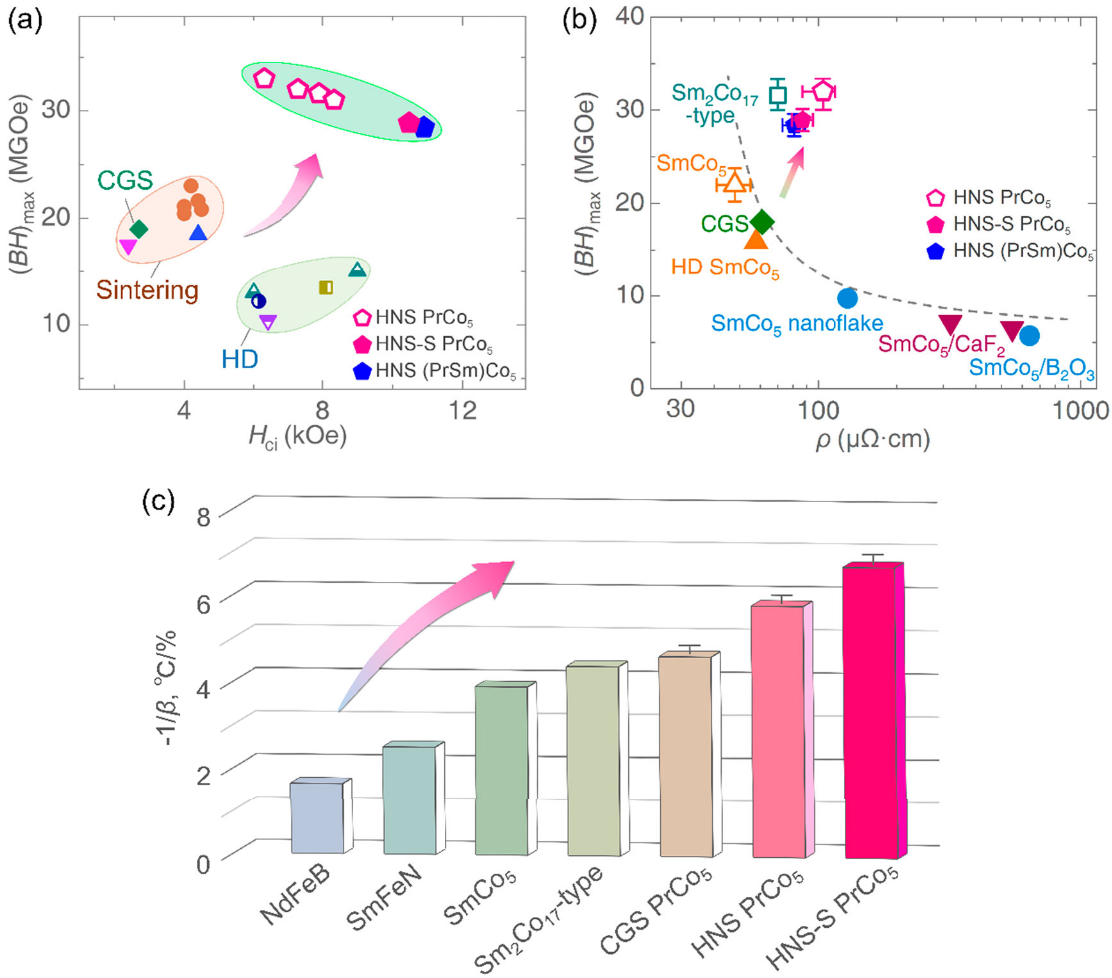

Figure 2. Multifunctionalities of new HNS $PrCo_5$ ferromagnets [10]. (a) Plot of energy density $(BH)_{max}$ and coercivity H_{ci} of the HNS materials. (b) Comparison of the $(BH)_{max}$ and electrical resistivity ρ. (c) Thermal stability of H_{ci} for various commercial magnets. The HNS ferromagnets exhibit an exceptional multifunctionality that combines multiple conflicting properties, including high $(BH)_{max}$, large H_{ci}, high ρ, and high thermal stability of H_{ci}.

the nano scale to atomic scale with abundant interfacial effects and atomic-scale composition variations. These features collectively resisted domain wall movement and electron transport in the material, ensuring its multifunctionality without sacrificing critical properties. This achievement demonstrates the superiority of hierarchical heterostructures in creating multifunctionality through overcoming multiple performance trade-offs with abundant interfacial effects and atomic-scale features.

To achieve the desired HNS, a Joule heating-based high-pressure constrained deformation technique was developed [10]. This method enables the rapid fabrication (within seconds) of fully dense bulk nanomaterials and allows for the controllable construction of nanoscale and atomic-scale structures. Using this technique, the HNS characteristics—such as grain size, SF density, and alignment degree—can be precisely modulated with processing parameters, yielding the bulk HNS with independently tailored nanoscale and atomic-scale features.

Prospects and challenges

The concept of HNS—a hierarchical heterostructure—is not limited to ferromagnetic materials. Although the proof-of-concept study made by the Zhang group [10] focuses on simultaneous control over domain wall movement and electron transport with abundant nano-scale interfacial effects and atomic-scale scattering of the HNS, the underlying concept can be extended to construct other multifunctional materials, allowing simultaneous control of phonon transport, light propagation, and the transport of reactants, as well as dislocation movement. As such, the design concept of hierarchical heterostructure can be applied to other functional and structural materials, providing a new paradigm for creating multifunctionalities. This strategy may enable significant advancements in energy efficiency, device reliability, and overall performance, as well as breakthroughs in performance limit of various materials, including thermoelectric, ferroelectric, and catalytic materials, in which various performance trade-offs exist [9]. Practical integration of these advanced materials into real-world applications, such as electric vehicles, aerospace actuators, and medical devices, will be a critical step. Collaborations with industry partners could facilitate the transition from laboratory-scale research to commercial products.

Although the success of the HNS strategy in $PrCo_5$ ferromagnets opens up new avenues for the development of multifunctional materials, creating such complex hierarchical heterostructures with independently tailored features remains a great challenge. New technologies need to be developed in order to overcome this challenge. The high-pressure constrained thermal deformation technique [10] provides a general platform for the controllable fabrication of bulk hierarchical nanomaterials. This groundbreaking technique can also be used to discover new metastable structures and phases that are unattainable with traditional methods. To achieve these goals, the relationship between processing conditions and the HNS substructure requires thorough investigation. The primary focus is to determine which factor—pressure, temperature, or time—serves as the key parameter in defining the HNS. Additionally, it is essential to examine how each of these parameters independently affects the complex microstructure of HNS. For example, the relationship between pressure and the alignment of grains and stacking faults, the impact of temperature on the stability of metastable structures, and the effect of processing time on the density of stacking faults and atomic-scale structural ordering require detailed analysis. Developing innovative strategies and technologies for the large-scale, rapid, and reproducible preparation of bulk HNSs is also critically important and remains a challenge. Compared to traditional microstructures, HNSs are more delicate and often incorporate metastable substructures, making their fabrication and control inherently more complex and challenging. Advanced techniques, such as 3D printing, may hold some potential for addressing these challenges. Moreover, establishing quantitative correlations between hierarchical heterostructures and various properties is also challenging and crucial for optimizing global properties of the multifunctional heterostructured materials. To reach this goal, the underlying mechanisms of the good combination of multiple conflicting properties in the heterostructures need to be revealed, where both experiments and theoretical simulations are required. However, simulating hierarchical heterostructures at multiple length scales, from atomic- to microscales, still remains a tough task, considering the computation power of current supercomputers. As such, new modeling approaches that connect these length scales need to be developed to fully capture the relationship between the hierarchical heterostructure and multifunctionality and to reveal the underlying mechanisms. All these are quite challenging tasks. But if we can overcome these challenges, the emerging hierarchical heterostructured materials with unprecedented multifunctionalities can be developed more easily.

In conclusion, the above example represents a significant leap in the field of multifunctional materials by engineering hierarchical heterostructures [10]. The HNS strategy not only addresses long-standing trade-offs in ferromagnetic materials but also sets the stage for future innovations in multifunctional materials. As we look to the future, the continued exploration and refinement of this new hierarchical heterostructure approach will hold immense promise for advancing technology and addressing global challenges in energy efficiency, resource conservation, and beyond.

Disclosure statement

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