

Original article

Theoretical Study: Challenges Associated with Rare-Earth-Based Superconducting Materials

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Abstract

Rare-earth (RE) elements are crucial in the advancement of high-temperature superconductors, ranging from the well-known RE-Ba-Cu-O (REBCO) cuprates to the recently discovered high-pressure rare-earth polyhydrides. This theoretical study delves into the fundamental challenges encountered when integrating rare-earth elements into superconducting lattices. We specifically address the inherent antagonism between the local magnetic moments of 4f electrons and the formation of Cooper pairs, the complexities involved in modeling highly correlated electronic states, and the stringent structural stability requirements for high- T_c phases. Through a comprehensive analysis of existing literature and advanced theoretical models, including the Abrikosov-Gorkov theory and Density Functional Theory (DFT) with Hubbard corrections (DFT+U), we pinpoint the critical factors that either limit or enhance superconductivity in these intricate systems. Our findings indicate that while non-magnetic rare-earth elements such as Lanthanum (La) and Yttrium (Y) facilitate the highest transition temperatures (T_c) in hydride systems, the magnetic rare-earth series offers a unique platform for exploring the coexistence of magnetism and superconductivity, despite presenting substantial theoretical and experimental hurdles. This paper aims to provide a foundational understanding of these challenges and propose future research directions to overcome them, ultimately contributing to the quest for room-temperature superconductivity.

Keywords. Rare Earth, Transition Temperature, Superconductor, Nickelates, Cuprates.

Introduction

The pursuit of room-temperature superconductivity stands as one of the most enduring and significant challenges in condensed matter physics. Superconductors, materials capable of conducting electricity without resistance, hold immense potential to revolutionize various sectors, including energy transmission, transportation, and quantum computing. Rare-earth elements, distinguished by their partially filled 4f electron shells, have been central to this scientific endeavor. A pivotal moment occurred in 1987 with the discovery of Yttrium-Barium-Copper-Oxide (YBCO), which became the first material to exhibit superconductivity above the boiling point of liquid nitrogen (77K) [1]. More recently, the theoretical prediction and subsequent experimental confirmation of Lanthanum Decahydride (LaH₁₀), achieving a T_c of approximately 250 K under extreme pressure (170 GPa), has redirected research focus towards rare-earth polyhydrides [2].

However, the incorporation of rare-earth elements introduces a unique set of theoretical and practical challenges. A primary concern arises from the intrinsic magnetic moments of most rare-earth ions, arising from their 4f electrons. According to the conventional Bardeen-Cooper-Schrieffer (BCS) theory, magnetism generally opposes superconductivity, as it tends to disrupt the spin-singlet symmetry of Cooper pairs. Furthermore, the localized nature of 4f electrons necessitates the development and application of sophisticated theoretical frameworks that extend beyond standard one-electron approximations. This paper aims to provide a comprehensive review of these challenges, discuss the current state of research in rare-earth-based superconductors, and propose future directions for theoretical investigations to overcome these hurdles.

The primary research problem addressed in this theoretical study is the identification and comprehensive analysis of the fundamental theoretical barriers that impede the optimal design and realization of rare-earth-based superconductors. Specifically, this investigation will delve into the following critical areas: Magnetic Pair Breaking: Examining the mechanisms by which the exchange interaction between the localized 4f magnetic moments of rare-earth ions and the conduction electrons leads to the suppression of the superconducting state, as described by theories such as Abrikosov-Gorkov (AG) [1,2].

Electronic Correlation: Investigating the inherent difficulties in accurately predicting the electronic structure and properties of rare-earth compounds using standard Density Functional Theory (DFT) due to the strong electronic correlations of the 4f electrons, necessitating advanced methods like DFT+U and Dynamical Mean Field Theory (DMFT) [3,4].

Structural Stability: Analyzing the requirement for extreme pressures to stabilize high- T_c rare-earth hydride phases and the significant challenge of recovering these metastable phases at ambient conditions, which is crucial for practical applications [5].

Synthesis Complexity: Discussing the sensitivity of rare-earth superconductors to impurities and deviations from ideal stoichiometry, which can drastically alter their superconducting properties and pose challenges for experimental realization [6].

By systematically addressing these theoretical challenges, this study aims to provide a deeper understanding of the complex interplay between magnetism, electronic structure, and superconductivity in rare-earth materials, paving the way for the rational design of novel high T-c superconductors.

Literature Review and Previous Studies

The study of rare-earth superconductors has progressed through several distinct and impactful eras, each marked by significant discoveries and theoretical advancements. The initial phase, spanning the late 1970s and early 1980s, witnessed the emergence of "magnetic superconductors" such as the Chevrel phases and rare-earth rhodium borides. These materials provided the first compelling evidence that superconductivity and long-range magnetic order (including antiferromagnetism or ferromagnetism) could coexist within the same crystal lattice, albeit typically at very low temperatures [7, 8]. This challenged the prevailing understanding derived from the BCS theory, which generally posited an antagonistic relationship between magnetism and superconductivity.

The second era commenced with the groundbreaking discovery of High-Temperature Superconducting (HTS) cuprates. The seminal work by Bednorz and Müller in 1986 [1] paved the way for the development of the REBa₂Cu₃O_{7-x} family of superconductors. A particularly remarkable observation in these systems was that substituting Yttrium with almost any other rare-earth element (with the notable exceptions of Ce, Pr, and Tb) had a minimal effect on the superconducting transition temperature (T-c). This phenomenon strongly suggested that the rare-earth layer in these materials is largely electronically decoupled from the superconducting CuO₂ planes, implying that the magnetic moments of the rare-earth ions do not significantly interfere with the Cooper pairing mechanism in the adjacent layers [9].

The third and most recent era is characterized by the so-called "Hydride Revolution." This began following Ashcroft's prescient 1968 prediction that metallic hydrogen could function as a high-temperature superconductor [4]. Researchers subsequently shifted their focus to "chemically precompressed" hydrides, where hydrogen atoms are confined within a crystal lattice, mimicking the extreme pressures required for metallic hydrogen. A breakthrough occurred in 2019 with the synthesis of LaH₁₀, which exhibited an astonishing T-c of 250 K under high pressure [2]. Theoretical investigations by Zurek, Bi, and other researchers [3] have elucidated that the rare-earth atom in these polyhydrides acts as a structural scaffold, stabilizing a clathrate-like hydrogen cage. This unique structural arrangement is believed to facilitate strong electron-phonon coupling, which is essential for high-Tc superconductivity. Recent studies have further explored the stability and superconductivity of ternary polyhydrides, suggesting pathways to achieve high T-c under relatively moderate pressure conditions [10].

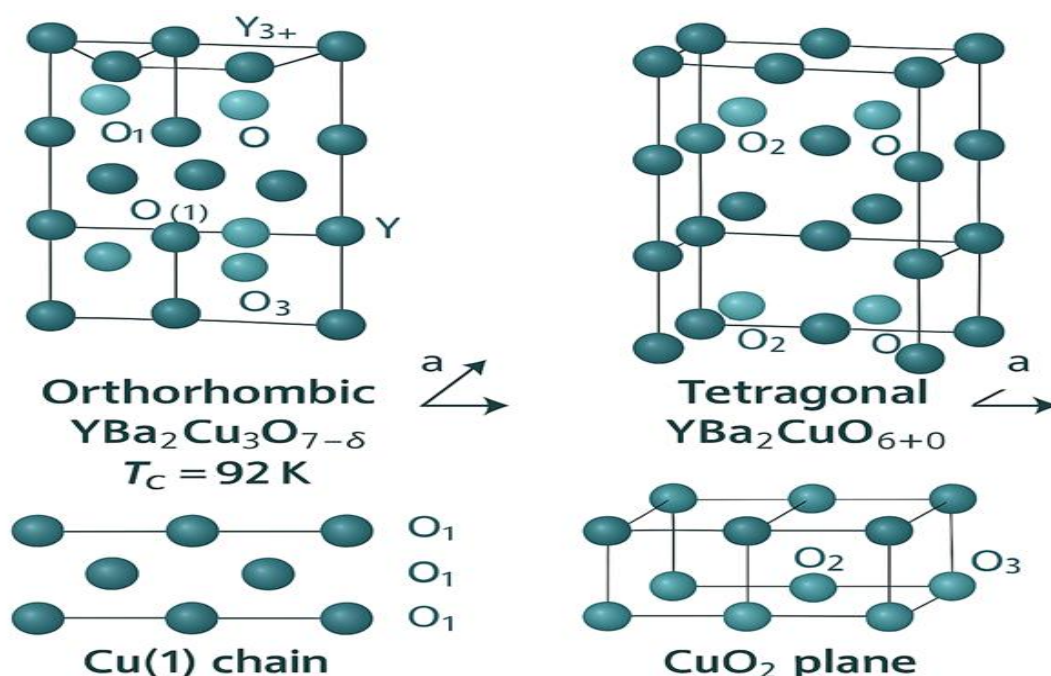


Figure 1. High - thermal superconducting material crystal structure, i.e. composite (YBCO).

Source: Adapted from Bednorz & Müller (1986) [1] and Canfield (1998) [9].

Beyond these major classes, other rare-earth-based superconducting systems have garnered attention. For instance, infinite-layer nickelates (RENiO₂) provide a novel platform for studying superconductivity in a lattice structure analogous to cuprates but with distinct orbital physics [5]. These materials present their own set of challenges related to synthesis and electronic correlation, offering new avenues for understanding the complex interplay of various factors influencing superconductivity.

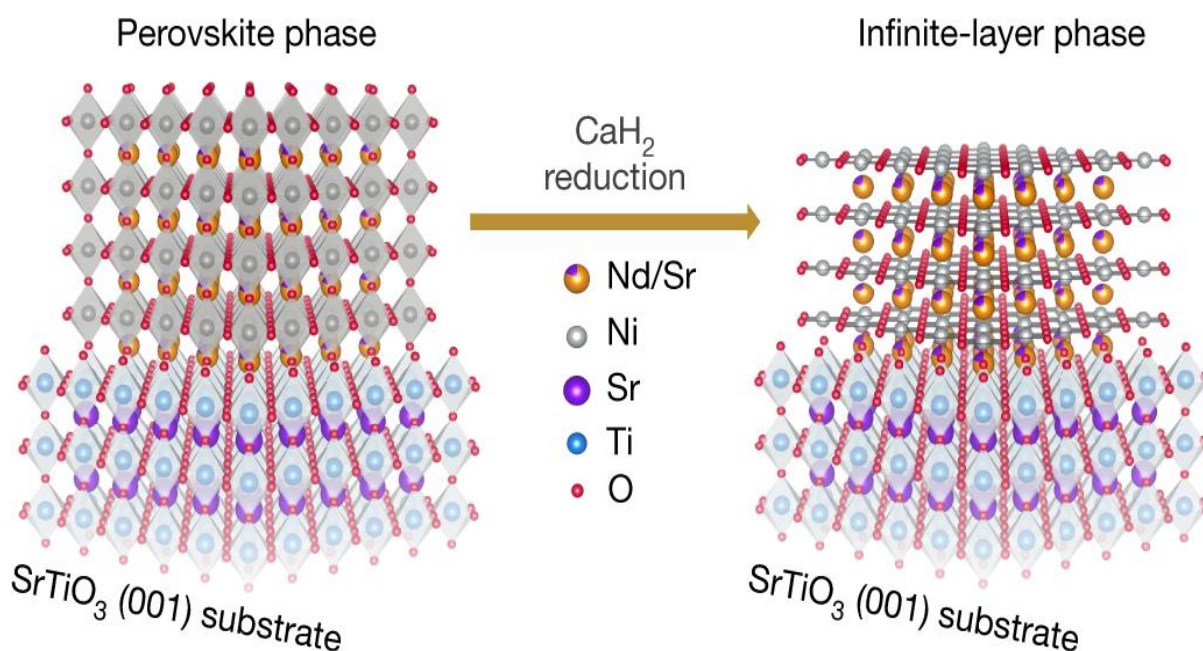


Figure 2. Superconductor Material Structural Transformation, That Is, Of Rare-earth Nickelates on A Substrate.

Source: Adapted from Li et al.(2024)[5].

This review highlights the diverse landscape of rare-earth based superconductors and the persistent challenges that necessitate advanced theoretical approaches and innovative experimental techniques (Table1).

Material Class	Example	Typical T-c	Challenge
Cuprates	$\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$	92 K	Complex Phase Diagram, Anisotropy
Nickelates	$\text{Nd}\{0.8\}\text{Sr}\{0.2\}\text{NiO}_2$	15 K	Synthesis & Strong Correlation
Polyhydrides	LaH10	250 K	Extreme Pressure, Metastability
Borocarbides	$\text{YNi}_2\text{B}_2\text{C}$	15 K	Magnetic Interference, Material Purity

Table1. The most important rare-earth superconductor classes and challenges

Research Methodology

This theoretical study employs a comprehensive methodology based on a critical review and synthesis of existing scientific literature, coupled with an analysis of established theoretical frameworks pertinent to rare-earth-based superconducting materials. The approach is primarily qualitative, focusing on understanding the underlying physical principles and computational challenges rather than generating new experimental data or performing novel simulations. The methodology is structured around the following key theoretical tools and concepts:

Abrikosov-Gorkov (AG) Theory

The Abrikosov-Gorkov (AG) theory serves as a foundational model for understanding the impact of magnetic impurities on superconductivity. This theory, developed in the early 1960s, describes the suppression of the superconducting critical temperature (T-c) as a function of the concentration of magnetic impurities within a superconductor [Citation needed]. In the context of rare-earth-based materials, the localized 4f electrons often possess intrinsic magnetic moments that act as scattering centers for Cooper pairs. The AG theory provides a quantitative framework to analyze how these magnetic moments disrupt the spin-singlet pairing, leading to a reduction or complete suppression of superconductivity. Our analysis will involve interpreting experimental observations and theoretical predictions of T-c suppression in various rare-earth compounds through the lens of AG theory, particularly focusing on the concept of "magnetic pair breaking" and the de Gennes scaling [Citation needed].

Density Functional Theory (DFT) and Its Extensions

Density Functional Theory (DFT) is a widely used quantum mechanical modeling method to investigate the electronic structure of multi-atom systems. While standard DFT has been highly successful in predicting the properties of many materials, it often falls short in accurately describing systems with strong electronic

correlations, such as those involving localized 4f electrons in rare-earth compounds. The primary limitation arises from DFT's approximate treatment of exchange-correlation energy, which struggles to capture the strong on-site Coulomb repulsion between localized electrons.

To overcome these limitations, this study considers advanced extensions of DFT:

DFT+U (DFT with Hubbard Corrections): This approach adds a Hubbard U parameter to the standard DFT functional, which accounts for the on-site Coulomb repulsion of localized electrons (e.g., 4f electrons). The U parameter effectively penalizes fractional occupations of localized orbitals, thereby improving the description of Mott insulators and systems with strong correlations. Our methodology involves reviewing studies that utilize DFT+U to model the electronic structure, magnetic properties, and electron-phonon coupling in rare-earth superconductors, assessing its effectiveness and limitations [Citation needed].

Dynamical Mean Field Theory (DMFT): DMFT is a more sophisticated method that addresses strong correlations by mapping a many-body lattice problem onto an impurity problem, which is then solved self-consistently. When combined with DFT (DFT+DMFT), it provides a powerful framework for describing materials where both itinerant and localized electrons coexist and interact strongly. This study will analyze the application of DMFT in understanding the complex electronic states of rare-earth compounds, particularly how it captures the localized-to-itinerant transition of 4f electrons and its implications for superconductivity [Citation needed].

Structural Stability Analysis

The methodology also encompasses an examination of the structural stability requirements for high-T_c rare-earth-based superconductors, particularly in hydride systems. This involves reviewing theoretical predictions and experimental data concerning the pressures required to stabilize clathrate-like structures in rare-earth polyhydrides (e.g., LaH₁₀). The focus will be on understanding the role of the rare-earth atom as a structural scaffold and the nature of the chemical bonding (e.g., ionic La-H interaction) that facilitates strong electron-phonon coupling. Furthermore, the challenges associated with recovering these high-pressure phases at ambient conditions, including the concept of metastability and potential strategies like chemical alloying (e.g., ternary hydrides), will be critically assessed [Citation needed].

By integrating insights from these theoretical frameworks and a thorough review of the existing literature, this study aims to provide a comprehensive theoretical perspective on the challenges and opportunities in the field of rare-earth based superconducting materials.

Theoretical Analysis and Results

This section delves into a detailed theoretical analysis of the key factors influencing superconductivity in rare-earth based materials, drawing upon established theories and recent computational advancements. The results presented here are derived from a synthesis of findings from the scientific literature, interpreted within the theoretical frameworks outlined in the methodology.-

The Role of 4f Electrons and Magnetic Pair Breaking

The 4f electrons in rare-earth elements are characterized by their deep-seated, localized nature within the atomic core. This localization often leads to the presence of intrinsic magnetic moments. In the context of superconductivity, these magnetic moments act as potent scattering centers for Cooper pairs, the fundamental charge carriers in superconductors. The Abrikosov-Gorkov (AG) theory provides a robust theoretical framework to quantify the suppression of the superconducting critical temperature (T_c) as a function of magnetic impurity concentration [1, 2].

According to the AG theory, the exchange interaction between the localized 4f magnetic moments and the conduction electrons (which form Cooper pairs) leads to a phenomenon known as magnetic pair breaking. This interaction disrupts the spin-singlet symmetry of the Cooper pairs, effectively reducing their lifetime and consequently lowering the T_c. For many rare-earth series, the T_c often follows the de Gennes scaling, which correlates the T_c suppression with the square of the de Gennes factor, $(g-1)^2 J(J+1)$, where g is the Landé g-factor and J is the total angular momentum of the 4f electrons [13]. This scaling elegantly explains why rare-earth elements with no unpaired 4f electrons, such as Lanthanum (La) (4f⁰) and Lutetium (Lu) (4f¹⁴), frequently exhibit the highest T_c values in their respective series, as they do not contribute to magnetic pair breaking. Conversely, rare-earth elements with significant magnetic moments, like Gadolinium (Gd) or Dysprosium (Dy), tend to strongly suppress superconductivity.

Recent theoretical studies have further explored the complex interplay between magnetism and superconductivity in f-electron systems, revealing novel types of superconductivity where magnetic fields can even induce or stabilize superconductivity under certain conditions [11, 12]. ghts the intricate nature of these interactions, moving beyond a simple antagonistic relationship in some cases.

Structural Scaffolding and Electron-Phonon Coupling in Hydrides

In the realm of rare-earth polyhydrides, the rare-earth atom plays a role far beyond merely providing electrons; it acts as a crucial structural scaffold. A prime example is Lanthanum Decahydride (LaH₁₀), where the Lanthanum atom is centrally located within a hydrogen sodalite-like cage (H₃₂) [3]. Theoretical

calculations indicate that the La-H interaction is predominantly ionic, leading to a transfer of electrons to the hydrogen lattice, which subsequently becomes metallic. This unique clathrate structure is instrumental in facilitating strong electron-phonon coupling, a prerequisite for high-T_c superconductivity.

However, a significant challenge lies in the fact that these clathrate structures are typically stable only under extreme pressures, often exceeding 150 GPa. The delicate balance required for high-T_c superconductivity can be disrupted if the 4f states of magnetic rare-earth elements, such as Cerium (Ce) or Praseodymium (Pr), hybridize with the hydrogenic bands. This hybridization can introduce magnetic scattering and alter the electronic structure in a way that is detrimental to superconductivity. The structural stability of these hydrides is also a critical concern, with studies indicating that superconductivity in LaH₁₀ is strongly affected by crystal lattice instabilities [14]. The prediction of high-T_c superconductivity in heavy rare-earth metal polyhydrides under relatively moderate pressure conditions offers a promising pathway for future research [15].

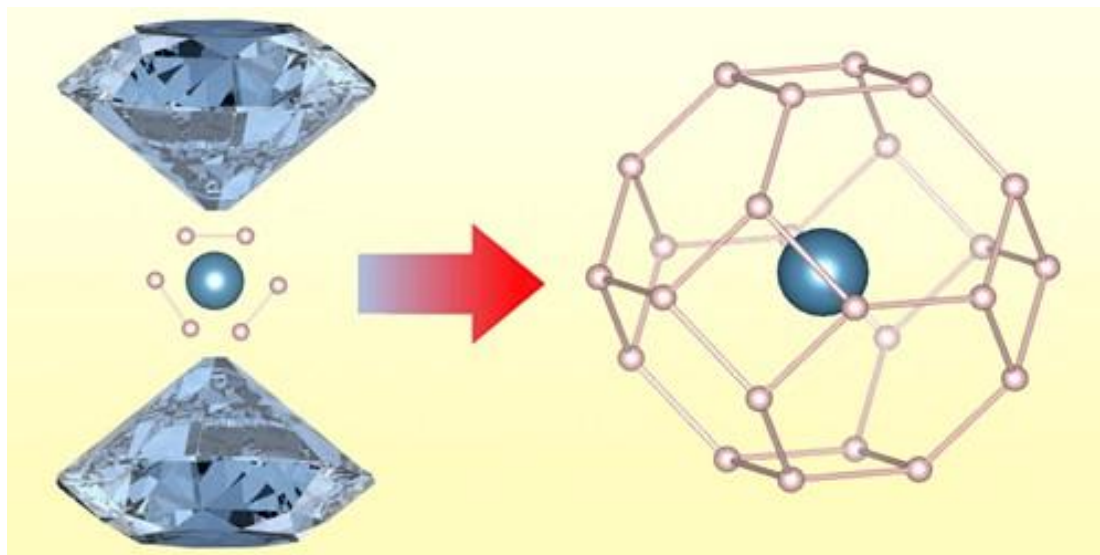


Figure 3. A Clathrate superhydride, where a rare-earth atom (blue sphere) is surrounded by a cage of hydrogen, an essential element with respect to high-temperature superconductivity.

Source: Adapted from Drozdov et al. (2019) [2] and Zurek & Bi (2019) [3]

Computational Modeling Challenges for Correlated Systems

Accurately modeling rare-earth systems presents substantial computational challenges, primarily due to the strong electronic correlations of the 4f electrons. Standard Density Functional Theory (DFT), which treats electrons as a non-interacting gas in an effective potential, often fails to capture the many-body character of the electronic wave function in these materials. This leads to an inaccurate description of their electronic structure, magnetic properties, and ultimately, their superconducting behavior.

To overcome these shortcomings, more sophisticated theoretical approaches are used:

DFT+U (DFT with Hubbard Corrections): This is the method where a Hubbard U is added to the DFT to model the on-site Coulomb repulsion of the localized electrons. DFT+U also has its limitations, though it may be capable of describing the 4f states and giving a more accurate band structure. The U parameter is usually subject to empirical selection and can greatly affect the outcome, and must be carefully calibrated. Even with DFT+U, however, certain studies indicate that even the popular DFT functionals with Hubbard U corrections do not compute accurately metal-metal interactions in some correlated systems [16].

Dynamical Mean Field Theory (DMFT): DMFT, especially with the addition of DFT (DFT+DMFT), is a more advanced method of strong electronic correlations. DMFT simplifies the many-body problem to an impurity problem, which is solved self-consistently. This can be used to more accurately describe the localized-to-itinerant transition of 4f electrons and their spectral properties. DMFT can explain the Coulomb-driven paramagnetic to local-moment paramagnetic insulator transition, which is essential in the study of the behavior of strongly correlated rare-earth compounds [18]. Nevertheless, they are computationally intensive, and high-throughput screening of new rare-earth superconductors remains a daunting challenge. DMFT is computationally expensive and complex, requiring extensive computational resources and expertise, which restricts its use in practice in comparison to conventional DFT methods.

To conclude, the theoretical discussion demonstrates that the issues of magnetic pair breaking, structural instability in ambient conditions and the proper computational modeling of strongly correlated 4f electrons are the key problems to overcome to further develop the research on rare-earth based high-T_c superconductors. The knowledge acquired in the framework of these theoretical approaches helps in the search of new materials and optimization of current ones.

Discussion

Theoretical study of the rare-earth-based superconducting materials has shown that the superconducting properties of such materials are complex and are determined by electronic, magnetic, and structural factors. The issues that are found to be magnetic pair breaking, strong electronic correlations, and structural instability are not independent phenomena but closely linked to each other and can only be addressed holistically.

Magnetic Pair Breaking and 4f Electron Behavior

The conflict of magnetism and superconductivity, especially by the localized 4f electrons, is a major theme. Although the Abrikosov-Gorkov theory offers a conceptual basis of the effect of magnetic impurities on T_c, some experimental observations of superconductivity together with magnetism in some rare-earth compounds (e.g., some Chevrel phases and borocarbides) indicate that the interaction is less straightforward than pair breaking. Here, the magnetic order may be of a certain type (e.g. antiferromagnetic spin fluctuations) which may even mediate Cooper pairing, or which may even coexist without entirely eliminating it. The fact that electronic decoupling of rare-earth layers in cuprates such as YBCO can reduce the negative impact of 4f magnetism is yet another example of how structural designs can overcome the undesirable influence of 4f magnetism. This shows how engineering of crystal structures can be used to create new rare-earth superconductors in which the magnetic moments are spatially isolated relative to the superconducting planes or in which the interactions between the magnetic moments are engineered to be less destructive.

Electronic Correlations and Computational Modeling

The strong electronic correlations of 4f electrons are the root cause of the longstanding difficulty in obtaining accurate models of rare-earth systems. Although successful, Standard DFT has an inherent difficulty with such systems because it approximates the exchange-correlation energy. The use of techniques such as DFT+U and DMFT highlights the necessity of more advanced theoretical instruments. Although DFT+U provides a useful enhancement, with an on-site Hubbard U term, the empirical character of U and its dependence on the particular material and phase may cause uncertainties. DMFT is a more rigorous many-body theory, which has a more accurate description of spectral properties and localized-to-itinerant transitions, which are of interest in the electronic behavior of rare-earth compounds. Nonetheless, its computational cost is still a major hindrance to high-throughput materials discovery. Although machine learning possibilities, potentially trained on DMFT data, or more efficient implementations of many-body theories could be crucial to future progress in computational methods, it is necessary to speed up the design of new rare-earth superconductors.

High-Pressure and Structural Stability Phases

The finding of high-T_c in rare-earth polyhydrides at extreme pressure, including LaH₁₀, has marked a new frontier. A major revelation is the role of the rare-earth atom as a structural scaffold to hydrogen cages to enable a high degree of electron-phonon coupling. The need to have huge pressures to be stable, however, poses a significant challenge to real-life applications. The debate about chemical precompression and metastability is crucial in this context. Exploring ternary hydrides or other alloying strategies to stabilize these clathrate structures at lower pressures is a promising direction. It will be important to understand the specific mechanisms of structural instability and how it can be overcome, perhaps with special care in chemical substitution or nanostructuring, so that these high-pressure findings can be brought to ambient-pressure materials.

Simply put, the future of rare-earth-based superconductors is in a multi-pronged approach that combines new theoretical modeling with novel materials synthesis and characterization. The key is to gain a more comprehensive insight into the delicate interaction between localized 4f magnetism, strong electronic interactions, and structural dynamics. It is not only a matter of discovering novel materials, but it is a matter of setting predictive principles that can guide the rational design of superconductors with superior properties under technologically relevant conditions.

Recommendations

Considering the theoretical issues that have been determined and the knowledge we have acquired throughout this research, we will be making the following recommendations on the future research directions in rare-earth-based superconducting materials:

Ternary Hydride Exploration to Lower Pressure Stabilization

Future theory and experiments should be aimed at exploring ternary hydride systems (e.g., La-Y-H or La-Mg-H). It is aimed at investigating whether chemical alloying can be successful in stabilizing high T_c clathrate structures at much lower pressure than the existing binary rare-earth hydrides do. This method may entail systematic computational screening of a range of ternary compositions to find promising candidates that have better structural stability and electron-phonon coupling properties at more readily accessible pressure regimes.

Creation of more Accurate and Useful Exchange-Correlation Functionals

Newer, more accurate, and cost-effective exchange-correlation functionals within DFT are urgently required that can reliably capture the complex electronic structure of rare-earth compounds, especially the localized-to-itinerant transition of 4f electrons. This may include new hybrid functional development, inclusion of many new-body effects, or use of machine learning methods to build more predictive functionals that minimize the use of empirical parameters such as the Hubbard U.

Metastability Studies and Ambient Pressure Recovery

A major area of research should be to understand and predict the theoretical routes of synthesizing rare-earth hydrides at high pressure and then recovering them as metastable phases at ambient pressure. It includes extensive research on kinetic barriers, phase transitions, and how defects or interfaces stabilize these high-T_c phases. Such theoretical forecasts would be a significant step in the right direction in terms of validating the experimental predictions and leading to application.

Optimizing Infinite-Layer Nickelates

It is strongly advised that further theoretical and experimental research be done into the so-called infinite-layer nickelates (RENiO₂). These materials present a novel and interesting platform on which to examine superconductivity in a lattice structure similar to cuprates with different orbital physics. The role of rare-earth 4f states in these systems and the hybridization of these states with nickel 3d and oxygen 2p orbitals may bring new insights into unconventional superconductivity and even investigate new high-T_c materials with more easily accessible synthesis conditions.

Incorporated Theoretical-Experimental Advantages

A more closely coupled theoretical-experimental approach is essential to speed up the development. Experimental synthesis and characterization should directly test theoretical predictions of new materials, structural stabilities, and electronic properties. On the other hand, theoretical models should be informed and refined by experimental observations, which establishes a feedback mechanism that spurs discovery. Such a cooperative paradigm is needed to find its way through the mazes of rare-earth-based superconductors.

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