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Research Progress on the 11 and 122 Systems of Iron-Based Superconducting Materials

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Abstract: Superconductivity was discovered 113 years ago in 1911. Due to the unique properties of superconducting materials, such as zero resistivity and perfect diamagnetism, they play a critical role in energy, healthcare, and other industries. As scientific research continues to advance, various types of superconducting materials, including iron-based superconducting materials, have attracted increasing attention from scientists. This paper provides an overview of the research progress and the significant development potential of various iron-based superconducting materials (primarily the 11, 122 and 1111 systems), with the aim of offering guidance and suggestions to researchers.

Keywords: iron-based superconducting materials, superconducting materials, research progress, development history

1. Introduction

In the ongoing exploration of exotic states of matter and the development of novel technologies, superconducting materials undoubtedly occupy a pivotal position. [1] Superconducting materials are a unique class of substances with special properties under specific conditions, including zero resistance (where the material's resistance disappears, allowing current to flow without loss) [2] and complete diamagnetism (where, in a superconducting state, the external magnetic field is completely expelled, resulting in zero magnetic induction intensity), among other characteristics. [3] Due to these exceptional properties, scientists have continuously explored various applications based on superconducting materials, enabling them to play a significant role in energy, healthcare, and other fields. [4]

In comparison with other superconducting materials, iron-based superconductors have attracted significant attention from scientists due to their excellent properties and great potential for applications. In 2008, a Japanese research group discovered 26 K superconductivity in $LaFeAsO_{1-x}F_x$, which garnered widespread international attention and marked the beginning of a new chapter in high-temperature superconducting materials. [5] However, there are few review articles on iron-based superconductors.

Therefore, this paper systematically introduces the development and modification methods of various iron-based superconducting materials in recent years, highlights the advantages and considerable development potential of these materials (mainly the 11 and 122 systems), and aims to

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provide guidance and suggestions to researchers to promote the research and development of high-performance superconducting materials.

2. The History of Superconducting Materials

In 1911, Dutch scientist Kamerlingh Onnes discovered that the resistivity of mercury (Hg) almost became zero at a temperature of 4.2 K, a phenomenon known as superconductivity. [6] After Onnes's discovery, many materials were tested for superconductivity, and rules of thumb were established to aid in the discovery of additional superconductors. It was soon found that only metals exhibited superconductivity, and poor conductors were better suited for superconductivity than good conductors. [7] The search for high-temperature superconductors is akin to navigating an ocean without an accurate compass; researchers must rely on theoretical guidance and intuition. [8] In this sense, the pursuit of room-temperature superconductors is a truly challenging subject and a typical research area. [9] As shown in Figure 1, which depicts a timeline of major events in the history of superconducting materials.

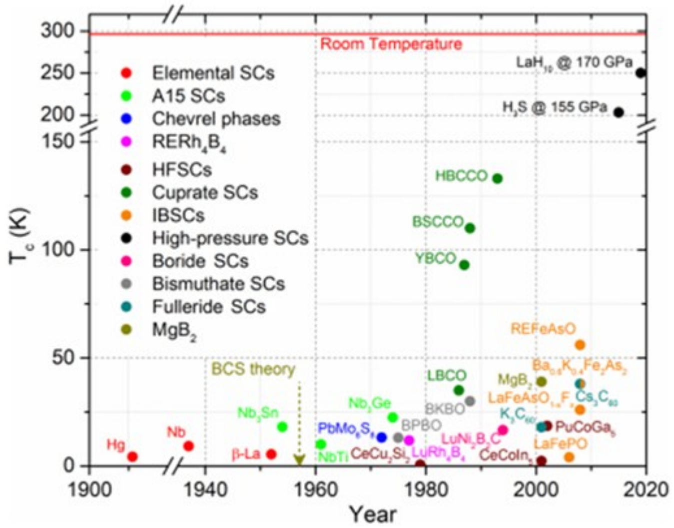

Figure 1: The History of Superconducting Materials

In the 1950s, Bardeen, Cooper, and Schrieffer developed the BCS theory [10], which successfully explained the electron pairing mechanism of superconductivity at the microscopic level. [11] The theory proposed that, at ultra-low temperatures, electron-phonon coupling results in a weak attractive interaction between electrons, leading to the formation of Cooper pairs. [12] Since Cooper pairs are bosons, they undergo Bose-Einstein condensation at low temperatures, creating a superconducting gap that enables electrons to move without resistance in metals. [13] The introduction of BCS theory marked a significant shift in our understanding of superconductivity, moving from a macroscopic to a microscopic perspective. It also concluded that the critical temperature of 40 K is the upper limit for various superconducting materials. [14] This theory not only explains the properties of conventional superconductors but also provides frameworks that have influenced many areas of physics, such as nuclear structure and Bose-Einstein condensate physics. The pairing concept, introduced by Leon Cooper, is an essential component of BCS theory, where lattice vibrations facilitate attractive interactions between electrons. [15] The measurement of isotopic effects on Tc stimulated further investigation into lattice vibrations, with theoretical studies by John Bardeen and David Pines providing compelling evidence for phonon-induced pairing interactions. [16] The multielectron nature of BCS theory evolved from the multibody wave function proposed by J. Robert

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Schrieffer. The details are well-documented elsewhere, and BCS theory has become one of the great theoretical breakthroughs of 20th-century physics. [17] According to BCS theory, under ultra-low temperature conditions, electron-phonon coupling results in weak attractive interactions between electrons, causing them to pair into Cooper pairs. Since Cooper pairs are bosons, they undergo Bose-Einstein condensation at low temperatures, which results in a superconducting energy gap, enabling the movement of electrons through the metal without resistance. [18] Over time, scientists recognized that while BCS theory had achieved significant success in conventional low-temperature superconductors, it was less applicable to the study of room-temperature superconductors. This limitation has introduced new challenges in the quest for room-temperature superconducting materials. Despite the continuous efforts of researchers to increase the critical temperature of superconducting materials, it remained stuck at 40 K for a long time. It wasn't until 1986 that German physicist Ponoroz and Swiss physicist Müller discovered that barium-lanthanum [19] superconductors could reach a critical temperature of 77 K. In 1987, Zhao Zhongxian and his team independently discovered a superconductor with a critical temperature of 93 K in the liquid nitrogen temperature range, sparking global excitement once again. [20] In comparison with other superconducting materials, iron-based superconductors have attracted significant attention from scientists due to their excellent properties and great potential for applications.

3. Iron-based Superconducting Materials

With the ongoing research by scientists, iron-based superconducting materials have gradually developed into a vast system, including the iron-based 11, 122 and 1111 systems, among others. In this section, we will systematically introduce the development process and modification methods of these materials. [21]

3.1. 11 System

The iron-based 11 system is a type of iron-based superconducting material with the simplest structure, consisting solely of a Fe_2X_2 layer (where X represents S, Se, or Te), composed of common FeX_4 units, and lacking a carrier reservoir layer. [22] Moreover, because the 11 system does not contain highly toxic or active metal elements, its safety and stability offer clear advantages compared to other systems. [23] Among the 11 systems, FeSe is the most widely studied. FeSe is the simplest known iron-based superconductor and has been the focus of intensive research, even after more than a decade of scientific investigation. [23] It is believed that the simplicity of the structure may provide valuable insights into how the mechanisms of superconductivity in these compounds can be elucidated. One of the most intriguing challenges is recovering the structure of the superconducting (SC) gap, which may be key to uncovering the interaction mechanisms and understanding the SC state. The most direct techniques for obtaining SC bandgap information are spectroscopic methods, such as angular-resolved photoelectron spectroscopy (ARPES), scanning tunneling spectroscopy (STS), and point-contact Andreev reflection (PCAR) spectroscopy. [24] Although FeSe has clear advantages within the iron-based 11 system, it still cannot surpass the temperature limit of 8K. However, after ongoing research, scientists have successfully increased this temperature limit to 15K by replacing Se with Te, resulting in the compound $FeTe_{0.55}Se_{0.45}$. With the advancement of scientific studies, researchers have used the liquid ammonia method to intercalate FeSe, leading to the development of $A_x(NH_3)_yFe_2Se_2$ (where A represents Li, Na, Ca, Sr, Ba, etc.) superconducting materials. Among these, $Na(NH_3)_yFe_2Se_2$ has a critical temperature as high as 46K. Additionally, FeSe's critical temperature was increased to 36.7 K through pressurization. In 2012, Xue's research group successfully observed a superconducting signal at 65 K by preparing a monolayer of FeSe on an STO substrate, and in

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subsequent experiments, they observed a superconducting transition exceeding 100 K. Although this high Tc is remarkable, the exact mechanism behind it requires further investigation.

3.2. 122 system

The 122 superconducting family primarily refers to superconducting materials formed by doping or pressurizing the Fe-As-based 122 compound $AeFe_2As_2$ (where Ae refers to alkaline earth or rare earth elements such as Ca, Sr, Ba, and Eu) as the base material. [25] Compared to other systems, it is easier to obtain high-quality single crystals from 122-system iron-based superconducting materials, making them a frequent subject of scientific study.

In 2008, German scientists discovered 122 families of iron-based materials with a Tc as high as 38K in $(Ba_{1-x}K_x)Fe_2As_2$, which garnered significant attention. The discovery of superconductivity in iron-phosphorus compounds in 2008 further spurred interest in the fundamental and applied research of superconducting materials. [26] Soon after, many other series of new compounds were identified. Among them, SmOFeAs (T_c = 55K, type 1111) and Sr/BaKFeAs (Tc = 38K, type 122) are particularly relevant for applications due to their high transition temperatures. As shown in the figure, $AeFe_2As_2$ has a tetragonal structure (I4/mmm) in a paramagnetic metallic state at room temperature and undergoes a first-order structural and magnetic phase transition into an orthorhombic structure with antiferromagnetic order (Fmmm) upon cooling. Doping or pressurization of the parent compound suppresses these magnetic and structural transitions, inducing superconductivity. [27]

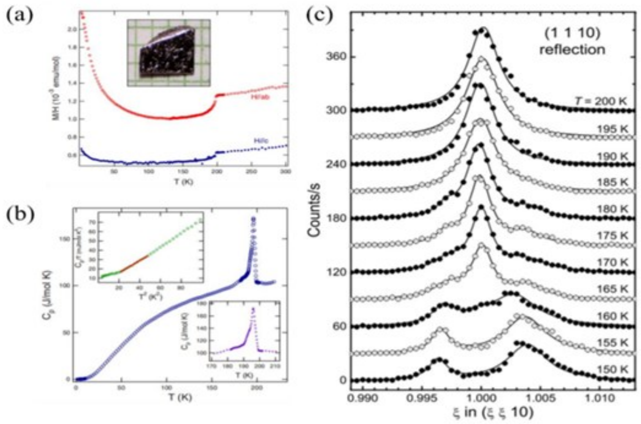

Figure 2: Magnetic and Structural Phase Transitions of $SrFe_2As_2$

3.3. 1111 system

Notably, the 1111-system compounds have garnered significant attention due to their exceptional properties. For instance, $SmFeAsO_{1-x}F_x$ boasts the highest critical temperature recorded at 57.5 K, accompanied by impressively high critical fields, estimated to exceed 200 T, and a critical current density surpassing 10^6 A/cm². However, the synthesis of novel 1111-system compounds remains a challenging endeavor, primarily because these phases are metastable and necessitate high-temperature synthesis methodologies under elevated pressure conditions.[28]

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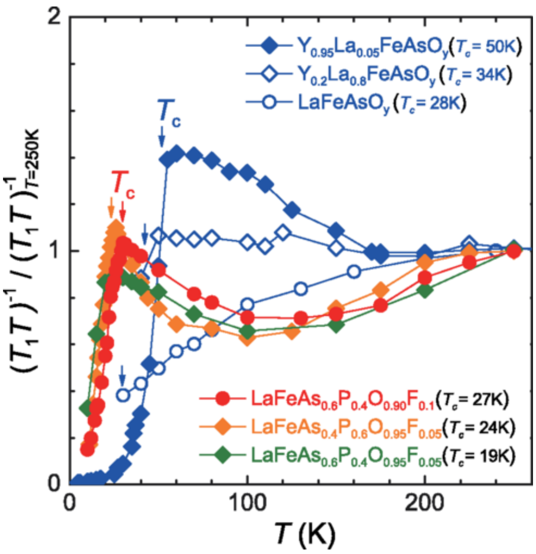

Figure 3: The NMR experiment result of $1/T_1T$ against T for $LaFeAs_{1-x}P_xO_{1-y}F_y$ and $Y_zLa_{1-z}FeAsO_y$.

In specific instances, the variation in T_c upon element substitution has been observed to exhibit non-monotonic behavior. This has, on occasion, given rise to speculations that the Cooper pairing mechanism in iron-based superconductors may involve multiple pairing mechanisms. A salient recent illustration of this phenomenon is found in the hydrogen-doped 1111 system.[29] Notably, substantial electron doping can be achieved by substituting oxygen with hydrogen in LnFeAsO (where Ln = Gd, Sm, Ce, La). Contrary to expectations, superconductivity has been demonstrated to persist even at an electron doping level of up to 40%. Particularly intriguing are the cases of LaFeAs(O,H) and SmFe(As,P)(O,H), where the phase diagram reveals a double-dome structure in response to electron doping.[30]

Further evidence of this complexity is provided by another example involving an isovalent doping 1111 system. In this case, arsenic is partially substituted with phosphorus in LnFeAs(O,F) (where Ln = Nd, Ce, La). As shown in Figure 3, it is well-known that increasing the phosphorus content leads to an enlargement of the Fe-Pn-Fe bond angle (or a reduction in the pnictogen height).[31] Based on the empirical trend in T_c observed by Lee et al., one would anticipate a monotonic decrease in T_c with increasing phosphorus content. However, this raises questions about the underlying pairing mechanisms in these materials, as the actual Tc behavior may deviate from such simple predictions. Thus, the exploration of these 1111-system compounds continues to offer valuable insights into the complex interplay of factors influencing superconductivity in iron-based materials.[32]

4. Conclusions and Prospects

This paper introduces the discovery of superconductivity, the development history of traditional superconducting materials, and the discovery of novel iron-based superconducting materials. It focuses on the development history and landmark achievements of different iron-based superconducting material systems (the 11, 122 and 1111 systems), providing guidance for the design of novel, high-performance iron-based superconducting materials. Although significant progress has been made in this field, challenges remain, including:

Room-temperature superconducting materials: The continuous development of new high-temperature superconducting materials, such as iron-based superconducting materials, and the ongoing improvement of the theoretical temperature of superconducting materials increase the possibility of room-temperature superconducting materials being used in various fields, including energy and medicine.

Rational design of materials: In the future, with the continuous advancement of artificial intelligence technology, the theories related to superconducting materials will be gradually refined. The combination of artificial intelligence technology and superconducting materials will inevitably make significant contributions to the progress of humankind.

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