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High-pressure study on some superconductors

K Q Li, Z A Ren, Y S Yao, G C Che and Z X Zhao

National Laboratory for Superconductivity, Institute of Physics, CAS, PO BOX 603, Beijing 100080, China

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Abstract

High-pressure study has played an important role in the investigation of conventional superconductors. Since the discovery of cuprate superconductors, high-pressure study has become even more important, especially as regards high-pressure synthesis and the effect of pressure. In this report, the new materials Ca-doped Pr-123, (Fe, Cu)-1212, and MgB₂—a very new and interesting system synthesized under high pressure with good quality—will be discussed.

Chemical inner pressure has been thought to explain the high T_c of Cadoped Pr-123. As another possibility, the replacement of the physical pressure effect by a chemical effect will be discussed.

1. Introduction

High-pressure study has played an important role in the investigation of conventional superconductors. This field still has great vitality and it has led to revision of the periodic table of superconductors. Many elements that are not normally superconducting become superconducting under high pressure. Very recently, Eremets *et al* [1] showed that under high pressure, boron becomes superconducting.

Since the discovery of cuprate superconductors, high-pressure study has become even more important, especially as regards high-pressure synthesis and the effect of pressure. In this report, the Ca-doped Pr-123 and $(Pr_{1-x}Re_x)$ -123 system, Fe-doped Y-123, and MgB₂—the very new and interesting system synthesized under high pressure with good quality—will be discussed.

2. High-pressure synthesis

2.1. Ca-doped Pr-123 and (Re, Pr)-123

The absence of superconductivity in the $PrBa_2Cu_3O_y$ (Pr-123) compound has attracted much attention. Many studies have been made to investigate why it is not superconducting and what suppresses the superconductivity in the (Re, Pr)-123 system [2]. Many theories and

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empirical rules have been proposed. The question remains open. Some authors have reported superconductivity in Ca-doped Pr-123 thin films with T_c about 50 K [3]. In our group, a high-pressure technique was applied to synthesize Ca-doped Pr-123 bulk materials [4]. By using a new synthesis route with a sintered precursor under inert gas and high-pressure synthesis in oxidizing conditions, a series of superconducting (Pr, Ca)-123 samples with different Ca contents were synthesized successfully [5].

The experimental results show that all samples with Ca contents from 0.3 to 0.7 prepared under high pressure exhibit a nearly single-phase tetragonal 123 structure. With Ca doping, all samples became superconducting. The onset T_c varies with Ca content; the highest T_c of about 115 K appears at a Ca content of 0.5. This is the highest T_c achieved in Re-123 compounds. The pressure coefficient measurement shows that T_c increases linearly with pressure at a rate of $dT_c/dP = +0.10$ K kbar⁻¹ with no sign of saturation up to 20 kbar. This is a rather big pressure effect.

We consider that the superconductivity in the (Pr, Ca)-123 system comes mainly from the substitution of Ca for Pr, which creates holes due to the difference in valence of Pr and Ca. The high solubility of Ca achieved by the high-pressure technique made it possible to induce enough carriers for superconductivity to appear. At ambient pressure, the solubility is only 0.3; no superconductivity was found [6]. The reason for the high T_c may be the new synthesis route and the chemical pressure induced by Ca insertion (see [5]).

A series of Ca-doped (Re, Pr)-123 (Re = La, Nd, Sm, Eu, Gd, and Y) samples, $(\text{Re}_{0.4}\text{Pr}_{0.6})_{0.5}\text{Ca}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_y$, were also synthesized under high pressure. The sample synthesis procedure was the same as that for the (Pr, Ca)-123 samples. Structure analysis shows that all samples crystallize in nearly single-phase tetragonal 123 structure. The dc susceptibility measurements demonstrate that all components exhibit superconductivity with T_c at around 100 K, no matter what the ion is [7]. This indicates that the content of Ca is a key factor in controlling carrier concentration and hence superconductivity in this system.

2.2. (Fe, Cu)-1212

The coexistence of magnetism and superconductivity has attracted a great deal of attention since this new class of material was found among the ruthenate cuprates, Ru-1212 [8] and Ru-1222 type [9]. Using the high-pressure synthesis technique, we demonstrated that a new Fe-containing 1212-type cuprate superconductor (Fe_{0.5}Cu_{0.5})SrBaYCu₂O_{7+ δ} ((Fe, Cu)-1212) also shows coexistence of ferromagnetism and superconductivity.

Samples were prepared by the conventional solid-state method, and then treated under high pressure (for details, see [10]). X-ray diffraction patterns indicate that these samples have identical structure types and are monophase. The structure is similar to that of Ru-1212. Fe ions substitute for the chain Cu and both oxygen basal plane sites are partially occupied.

The sample synthesized at ambient pressure was not superconducting and exhibited paramagnetic behaviour, while sample treated under high pressure had bulk superconductivity with T_c (onset) = 77 K. At the same time, both the resistance measurement and some typical isothermal magnetization loops for a high-pressure sample show that the sample has weak ferromagnetism at 150 K. This shows clearly the coexistence of ferromagnetism and superconductivity. The ferromagnetism might be caused by Dzyaloshinsky–Moriya-type superexchange interaction of Fe–O–Fe and Cu–O–Cu in (Fe, Cu)–O planes [10], which cause the spins to cant out of their original directions. As a result, the D–M-type superexchange interaction forms the FM state.

Further experiments such as annealing and magnetization studies indicate that the occurrence and disappearance of ferromagnetism and superconductivity are determined

by the oxygen contents and local structure of the samples, and the ferromagnetism and superconductivity are not caused by phase segregation.

In the conventional solid-state reaction method, when Fe is doped into Y-123 and the content exceeds 0.3, Fe ions will occupy the planar Cu sites and destroy superconductivity. In our experiment, the inert-gas reduction and high-pressure treatment prevent Fe ions from entering the planar Cu sites and, in principle, all Fe ions should occupy chain Cu sites. That is one reason for the superconductivity in this system.

2.3. MgB_2

The recent discovery of superconductivity with $T_c = 39$ K in the intermetallic compound MgB₂ [11] has generated great interest as regards both fundamental mechanisms and practical applications. Here we report on the superconducting properties of MgB₂ samples synthesized in two different ways, at ambient pressure and at high pressure. The results are compared [12].

The samples synthesized under high and ambient pressure are both single phase, and each has a superconducting transition at about 40 K. The high-pressure sample has a much better ZFC transition which reflects the good connectivity between grains and much stronger pinning. The bulk critical current density (J_c) for the high-pressure sample, 6.7×10^5 A cm⁻² at 6 K with no field, is 30 times that for the ambient-pressure sample. Also, the high-pressure sample is much denser (2.56 g cm⁻³) than the ambient-pressure one. Additionally, high pressure can overcome the difficulty in synthesizing MgB₂ under ambient pressure that Mg is extremely volatile at elevated temperature. These comparisons clearly show that the high-pressure synthesis technique is favourable for obtaining high-quality samples of this new type of superconductor.

3. Discussion

It is well known that a lot of high- T_c materials have a positive pressure effect coefficient. That is, T_c increases with pressure. The effect of pressure on the structure is the shortening of all cation–anion bond lengths. So the stabilization of the structure responsible for these higher T_c -values at ambient pressure might prove to be very rewarding. The substitution of small ions for large ions would induce a shortening of the lattice parameter. The effect of this chemical substitution should be equivalent to that of the external pressure—that is, the application of chemical inner pressure. However, the case is not in fact so simple. Substitution of Sr for Ba caused different effects on T_c in $(La_{1-x}Ba_x)_2CuO_4$ [13] and in Hg-1201 [14]. This indicated that the structural change caused by chemical substitution may not be the same as that induced by mechanical pressure. The *in situ* high-pressure studies carried out together with *in situ* structural studies may unveil which cation or anion substitutions produce, at ambient pressure, the structure stable under pressure and responsible for the higher T_c . That may lead to the obtaining of optimized materials by means of appropriate chemical substitutions, and increase of T_c .

The results for Hg–Re 1223 [15] shows that the decrease in T_c with chemical internal pressure may arise from the increase in the distance between two CuO₂ planes. In the Cadoped Pr-123 series, Ca²⁺ is smaller than Pr³⁺. So this substitution may cause the distance between the two CuO₂ planes to decrease, and this may be one possible reason for the higher T_c in (Pr, Ca)-123.

In general, solving the problem of 'chemical internal pressure' is quite helpful for understanding the origin of the highest T_c and the mechanism of high- T_c superconductivity. Also, there is a possibility of replacing physical pressure and reaching higher T_c at ambient pressure.

4. Summary

High-pressure synthesis is a powerful technique in the search for new superconducting materials. It favours the formation of metastable phases, increases the solubility of the substituted ion, and prevents some kinds of disorder. Thus under high pressure, many materials that do not exist at ambient pressure can be synthesized and materials that do not superconduct can be made superconducting. High pressure also has a great influence on the study of the physics and mechanisms of superconductors. During the search for new materials, many new physical phenomena have been discovered and new mechanisms were also proposed to account for them. High pressure is becoming more and more important in the development of high-temperature superconductivity science and technology.

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