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Au-doping effects in the $Mg_{1-x}Au_xB_2$ series

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Abstract

A series of Au-doped MgB_2 superconductors with different doping levels were prepared by means of solid-state reaction. X-ray diffraction analysis indicates that $Mg_{1-x}Au_xB_2$ samples with $x \leqq 0.236$ can still be identified as having a hexagonal structure, with a different space group from the pure MgB_2. The resistive measurements show that the addition of Au to MgB_2 results in a Tc decrease. The dependence of the crystal structure and the superconducting transition temperature on the doping concentration is discussed.

Keywords: MgB_2; Au-doping; Superconductivity

1. Introduction

Since the discovery of the medium Tc (~ 39 K) superconductor MgB_2 in 2001 [1], there have been quite a lot of reports on its synthesis, doping, phase formation and superconductivity mechanism, from basic investigations to application studies [2–7]. Unlike high temperature oxide superconductors (HTS), MgB_2 is likely to be a "conventional" superconductor. Its Tc is the highest known transition temperature for a non-copper oxide bulk material.

MgB_2 has a simple chemical composition and occurs in the so-called AlB_2 structure, which consists of alternating hexagonal layers of Mg atoms and honeycombed B atoms [1]. In HTS materials, chemical doping has been widely used to investigate the effect of electron and structural changes on the physical properties. Chemical doping in MgB_2 is done in different groups and various results have been presented. Doping with some elements, such as partial substitution of Al or Li for Mg leads to the decrease or loss of superconductivity [8,9]; some elements cannot be doped into the lattice and thus have no effect on Tc, as reported for Be-doped MgB_2 [10]. Zhao et al. [7] reported another type of doping effect, the dopant Ti yields nonsuperconducting phases without decreasing the Tc of the superconductor matrix and fine particles of Ti-rich phase may act as pinning centers. The resulting improvement of critical current density in MgB_2 superconductors by Zr doping has been presented [11].

Based on the similar radius of Au and Mg atoms and electron structure calculations by means of the BCS mechanism, we selected Au as the first substitution element for Mg and successfully obtained superconducting $Mg_{0.854}Au_{0.146}B_2$ samples with Tc ~ 37 K. We then proceeded to prepare a series of Au doped MgB_2 compounds. In this paper we present the preparation of the series $Mg_{1-x}Au_xB_2$ ($x = 0-1$) and their superconducting properties. The change of the superconducting transition temperature of $Mg_{1-x}Au_xB_2$ series with different substitution concentrations is also discussed.

2. Experimental

The $Mg_{1-x}Au_xB_2$ samples were prepared by a direct solid state reaction of the elements Mg, Au and B. According to a stoichiometric $Mg_{1-x}Au_x$/B ratio of 1:2 with x ranging from 0 to 1, the starting materials magnesium powder (Mg≥95%), gold powder (Au 99.99%) and boron powder (B 99.999%) were mixed, slightly ground and then pressed into pellets. In order to minimize the volatilization of Mg, the pellets were wrapped in a Ta foil, which was sealed in an evacuated quartz ampoule and sintered for 5 h at 800 °C before cooling to room temperature. The reaction temperature was based on the Au–B phase diagram [12]

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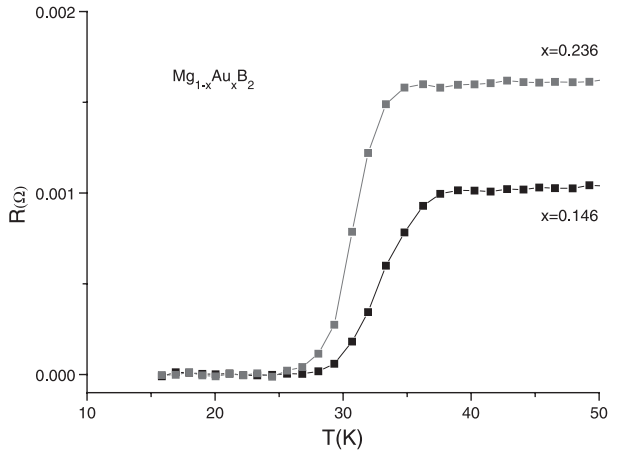

Fig. 1. Temperature dependence of the resistance of superconducting $Mg_{1-x}Au_xB_2$ samples.

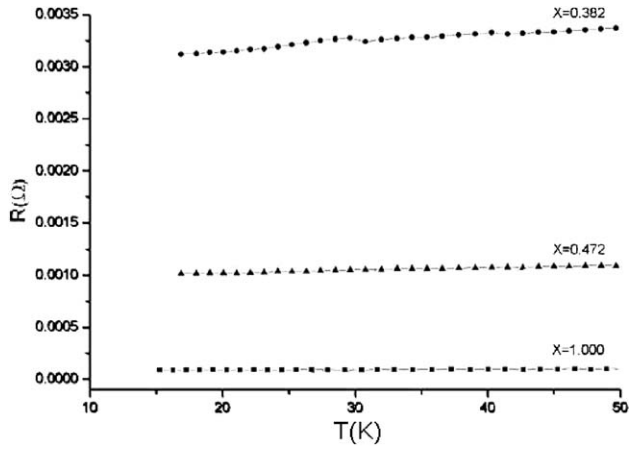

Fig. 2. Metallic $R-T$ dependence of the Au-doped $Mg_{1-x}Au_xB_2$ with $x \geqslant 0.382$.

and on the studies of thermodynamics of the Mg–B system [13]. Under these reaction conditions, we obtained a superconducting $Mg_{0.854}Au_{0.146}B_2$ sample with Tc ~ 37 K. We then made improvements on the reaction conditions, special care was taken to decrease the heating rate in the range of 650–800 °C because of the high volatilization of magnesium. Using the golden section principle, a sample series of $Mg_{1-x}Au_xB_2$ with $x = 1$, 0.618, 0.472, 0.382, 0.236, 0.146 (nominal composition) was prepared.

Phase identification was carried out by X-ray diffraction (XRD; 2θ range of 10–80°, with an X'pert MRD diffractometer, Philips, using Cu Kα radiation). The transition temperature was measured by a dc four-probe method using a cooling apparatus which can lower the temperature down to ~ 15 K. Microstructural observations were made by scanning electron microscopy (SEM; Amray-1910 E microscope at an acceleration voltage of 20 kV).

3. Results and discussion

All Au-doped products $Mg_{1-x}Au_xB_2$ (with $x \neq 1$) are black and appear to have a higher density and hardness than the pure MgB_2 sample. It has been found that the samples become denser with increasing doping content. For the maximal doping level where $x = 1$, i.e. in the case of the pure binary boron–gold system, we have obtained a bulk sample with considerable compactness. It seems that the high volatilization of Mg at experimental temperatures leads to porous products of low density. Although some techniques, including high-pressure sintering and proton irradiation, can be used to improve the samples quality [14], they suffer from the technical problems and are not suitable for the fabrication of MgB_2 wires and tapes. However, the chemical doping was found to be easily controlled.

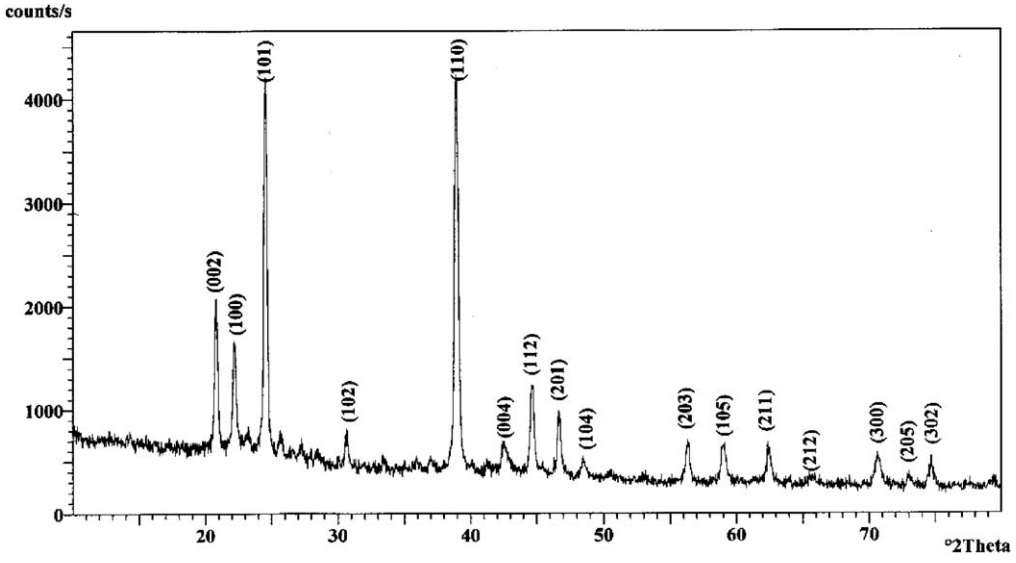

Fig. 3. Powder X-ray diffraction pattern of $Mg_{0.854}Au_{0.146}B_2$.

The results of the resistance–temperature ($R-T$) measurement indicate that the obtained Au-doped products with $x=0.146$ and 0.236 have superconductivity. As showed in Fig. 1, the onset of the transition for $Mg_{0.854}Au_{0.146}B_2$ and $Mg_{0.764}Au_{0.236}B_2$ occurs at temperatures of 37.6 and 35 K, respectively. The $R-T$ transitions for Au-doped samples remain sharp, but show a broader spar than the un-doped MgB_2 [1]. So zero resistance did not occur until about 25 K (25.6 K for $x=0.146$ and 24.5 K for $x=0.236$). The $Mg_{1-x}Au_xB_2$ samples have a lower Tc than the pure MgB_2. This decrease in Tc is in accordance with the BCS mechanism, since the substitution of a heavy atom Au in the site of a light atom Mg is expected to decrease the phonon frequency.

Fig. 2 exhibits metallic $R-T$ dependence for samples with higher doping levels ($x\geqslant 0.382$). No superconductivity was observed up to 15 K (the temperature limit of the used cooling apparatus), though the room-temperature resistance of these samples is remarkably low (below 1 Ω). It is found that the resistance decreases when the Au-doping concentration increases, which just reflects the metal properties of the element gold (Fig. 2). Au atoms may be not be introduced into the lattice of MgB_2 for higher doping levels ($x\geqslant 0.382$) and therefore no new phases are formed.

The X-ray diffraction pattern for $Mg_{0.854}Au_{0.146}B_2$ shows that all the intense peaks can be indexed assuming a hexagonal unit cell with $a=4.612$ and $c=8.525$ Å (Fig. 3). The similar X-ray analysis result is obtained for the sample with $x=0.236$. In the X-ray diffraction pattern for samples with $x=1$, one can only distinguish peaks from the cubic Au (Ref. 1996 JCPDS: 04-0784), i.e. the Au element did not react with boron. As shown in Fig. 4, the peaks from the XRD analysis of the non-superconducting $Mg_{1-x}Au_xB_2$ ($1>x\geqslant 0.382$) could not be attributed to pure phases, though

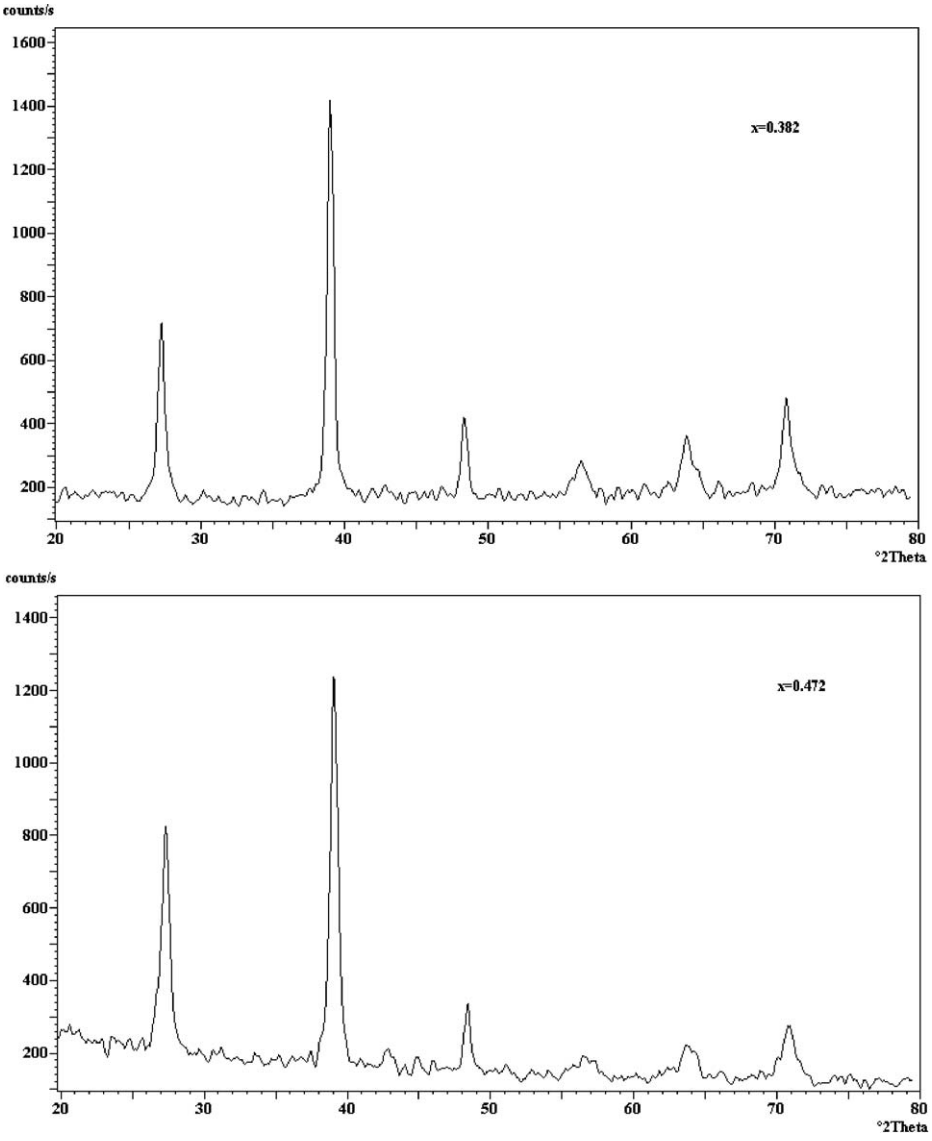

Fig. 4. XRD of $Mg_{1-x}Au_xB_2$ with $x=0.382$ and 0.472.

there exist some similar peaks to that of the low doping samples. The preparation of the doped samples by improving the reaction conditions is yet in progress. With further experimental results, we hope to explain the effects of Au-doping in $Mg_{1-x}Au_xB_2$ on the crystal structure and the resulting superconducting properties. Examination by electron microscopy indicated that the particle sizes for $Mg_{1-x}Au_xB_2$ powders were very small (the average grain size is smaller than ~ 1 μm), however, almost no regular crystalline grains were observed.

4. Conclusion

Au-doping can change the superconducting transition temperature of MgB_2 bulk. $Mg_{1-x}Au_xB_2$ samples with $x \leqslant 0.236$ prepared by solid state reaction have a lower Tc than the pure MgB_2. The substitution of a heavy atom (Au) in the site of a light one (Mg) is expected to decrease the phonon frequency and, therefore, to decrease Tc, which is consistent with the BCS mechanisms. For $x \geqslant 0.382$ no bulk superconducting transition occurs.

Acknowledgements

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