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Ni substitution effect on structure and superconductivity properties in $Li_2(Pd_{1-x}Ni_x)_3B$ boride system

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Abstract

The crystal structure and basic superconductivity parameters in $Li_2(Pd_{1-x}Ni_x)_3B$ are systematically studied by means of X-ray diffraction method, transport properties and magnetization measurement. The limiting solubility of Ni substitution at Pd site was less than 0.2. The lattice parameter a and superconducting transition temperature T_c decrease gradually with increasing Ni content x. Low critical field H_{c1} and upper critical field H_{c2} were estimated from the isothermal magnetization (M–H) measurements, and the results showed a decreased tendency with increase in x. From $H_{c1}(0)$ and $H_{c2}(0)$, the coherence length $\xi(0)$, penetration depth $\lambda(0)$, Ginzburg–Landau parameter $\kappa(0)$ and thermodynamic critical field $H_c(0)$ were estimated and the results are compared with the related systems. The present results supported that $Li_2(Pd_{1-x}Ni_x)_3B$ system is a conventional type-II superconductor.

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Keywords: $Li_2(Pd_{1-x}Ni_x)_3B$ system; Superconductivity parameters; Solid solution

1. Introduction

The recent discovery of superconductivity below 7–8 K in the ternary lithium borides, Li_2Pd_3B, has attracted a wide attention for its rich physical properties [1,2]. This compound is known to crystallize in a perovskite-like cubic structure (space group of $P4_232$) consisting of distorted Pd_6B octahedral [3], which is structurally similar to the superconductor $MgCNi_3$ [4], and also similar to the high-T_c copper oxide superconductors where the key structure is the oxygen-containing octahedra in some sense. Most likely, this system provides a model example bridging unconventional superconductivity with the classic BCS superconductivity. The theoretical and experimental (NMR measurement and photoemission experiment) results suggested that the superconductivity in Li_2Pd_3B is due to Pd 4d-electrons and these d-electrons have a strong coulomb correlation, dominating the electronic properties of this system [5,6]. As well as in other superconductors, elemental substitution serves as a useful way to modify the structure and other physical properties of Li_2Pd_3B system to study the underlying mechanism of superconductivity. Badica et al. [7] reported the basic superconducting parameters for pseudo-binary $Li_2(Pd_{1-x}Pt_x)_3B$ ($x = 0$–1) system. The results suggested that the observed superconductivity was of bulk type and the superconductivity parameters were similar to those of conventional type-II superconductors. Recently, it was indicated that the spin-triplet superconducting state was due to the broken inversion symmetry in Li_2Pt_3B by B^{11} and Pt^{195} NMR measurements [8]. This behavior is in contrast to those in the isostructural Li_2Pd_3B which is a spin-singlet superconductor [9]. In addition, Mani et al. [10] reported the lattice parameter and superconductivity transition temperature T_c for $Li_2(Pd_{1-x}Ni_x)_3B$ system. In this paper, to further understand the physical properties of $Li_2(Pd_{1-x}Ni_x)_3B$,

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the lattice parameter, superconducting transition temperature T_c, together with low critical field H_{c1}, upper critical field H_{c2}, coherence length $\xi(0)$, penetration depth $\lambda(0)$, Ginzburg–Landau parameter $\kappa(0)$ and thermodynamic critical field $H_c(0)$ are reported.

2. Experimental

Polycrystalline samples of $Li_2(Pd_{1-x}Ni_x)_3B$ were prepared by a two-step arc melting method under argon atmosphere. In the first step, alloys of $(Pd_{1-x}Ni_x)_3B$ ($x = 0$–1) were melted from the stoichiometric amounts of high-purity Pd, Ni and B on the water-cooled copper hearth. The total weight loss of this step was negligible, which was less than 0.3%. In the second step, Li was introduced with additional 10–40% to compensate the unavoidable loss of it during arc melting. The second procedure was repeated several times and the chosen samples which have the stoichiometric proportion mostly close to the $Li_2(Pd_{1-x}Ni_x)_3B$ by measuring the weight gain combined with the results of X-ray powder diffraction experiment (XRD). The samples were cut to rectangular for measurements of electrical resistivity, which were made using a standard four-probe technique in the temperature range from 1.8 to 280 K conducted by PPMS system (Quantum Design). The magnetization measurements were done using an SQUID (Quantum Design MPMS) in the temperature range from 2 to 30 K and the DC magnetic fields with a range of 0–6 T. The results are well repeatable.

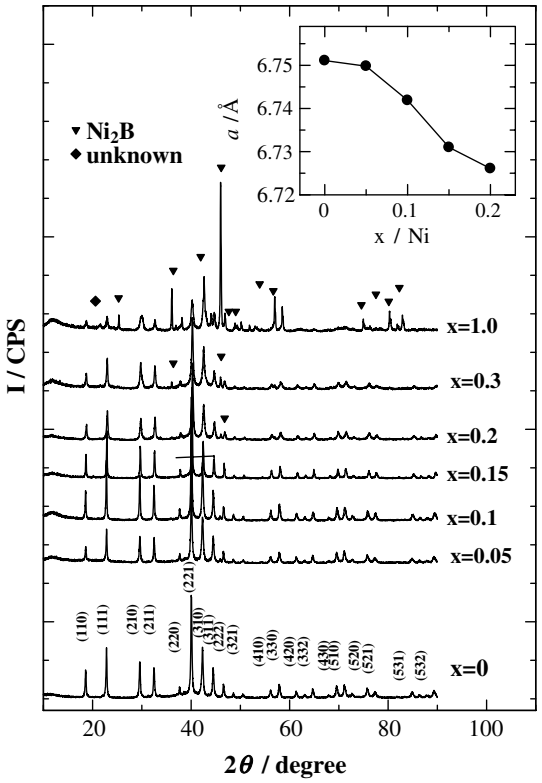

Fig. 1. Powder XRD patterns for $Li_2(Pd_{1-x}Ni_x)_3B$ ($x = 0$–1) system. The inset is the lattice parameter a as a function of x for $Li_2(Pd_{1-x}Ni_x)_3B$ ($x = 0$–0.2) system.

3. Results and discussion

The XRD spectra were performed on all of the $Li_2(Pd_{1-x}Ni_x)_3B$ samples and the results are shown in Fig. 1. For $x < 0.2$, no impurity peaks were found within the experimental errors, all samples have single phase with a cubic crystal structure belonging to $P4_232$ space group. When $x \geqslant 0.2$, some Ni_2B and unknown phases were found. It was found to be rather difficult to synthesis the Li_2Ni_3B sample by the present method. So, the limiting solubility of Ni substitution at Pd site in $Li_2(Pd_{1-x}Ni_x)_3B$ system was in the range of 0.15–0.20. The inset of Fig. 1 gives the change of lattice parameter a with x in $Li_2(Pd_{1-x}Ni_x)_3B$ ($x = 0$–0.2) system, which is calculated by the least-square method from the powder XRD data. It can be seen that the lattice parameter a gradually decreases with increasing x, which is similar to that of previously reported $Li_2(Pd_{1-x}Pt_x)_3B$ ($x = 0$–1) system [7] with the increase of substitution concentration and is consistent with Mani et al.'s [10] reported results.

Fig. 2 shows the temperature dependence of normalized electrical resistivity $\rho(T)/\rho(10\text{ K})$ and magnetization $M(T)/M(10\text{ K})$ (under field cooling mode) for $Li_2(Pd_{1-x}Ni_x)_3B$ ($x = 0$–0.2) system from 5 to 9 K. All the samples show a sharp superconducting transition. The superconducting transition temperature T_c was summarized and shown in the inset of Fig. 2. Apart from small differences in values (which can be attributed to different preparation conditions), the regularity of the T_c dependence on x was consistent with Mani et al.'s [10] reported results by temperature dependence AC susceptibility. This fact suggests that the quality of our samples and present results are highly reliable. Generally, T_c decrease systematically with increasing Ni substitution content of x. According to Mani et al. [10] the decrease in T_c is possibly due to the decrease in the density of state $N(0)$ and the increase in the average characteristic phonon frequency. Mochiku et al. [11] studied the distortion of $(Pd, Pt)_6B$ octahedron which was estimated by the quadratic elongation, $<\lambda>$, and the bond angle variation, $<\sigma^2>$, from the results of the refined neutron powder diffraction. They found the distortion of the Pt_6B octahedron was larger than that of Pd_6B octahedron for $Li_2(Pd, Pt)_3B$ and considered that this was the main reason for the Li_2Pt_3B to have lower T_c than that of Li_2Pd_3B. So, for the present Ni substituted $Li_2(Pd_{1-x}Ni_x)_3B$ system, the change of structure which has a close relationship with the distortion of Pd_6B octahedra is another reason for the decrease in T_c. According to Mochiku et al. [11], the (Pd, Pt)–B bond length which could be reflected by the lattice parameters, a, has a close relationship with the distortion of $(Pd, Pt)_6B$ octahedron. Despite from the present results, we cannot estimate the distortion of $(Pt, Ni)_6B$ octahedron precisely, the continuous and obvious decrease in a may indicate that the distortion of $(Pt, Ni)_6B$ octahe-