Superconductivity in the noncentrosymmetric Li$_2$Pd$_{3-x}$Ag$_x$B with $x = 0.0, 0.1$ and $0.3$

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Abstract

We studied the partial substitution of Ag in the noncentrosymmetric superconducting compounds Li$_2$Pd$_{3-x}$Ag$_x$B with $x = 0.0, 0.1$ and $0.3$. Magnetization, resistivity and specific heat measurements were performed in this system. From electrical measurements, we obtained the critical temperature $T_c$. The upper critical field at zero temperature was determined with the Werthamer–Helfand equation, $\mu_0 H_{c2}^{\text{WHH}}(0)$, and the coherence length, $\xi(0)$. The value of $\mu_0 H_{c2}^{\text{WHH}}(0)$ and $\mu_0 H_{c2}^{\text{linear}}(0)$ is lower than the calculated paramagnetic Pauli limit suggesting that the spin-triplet pairing is weak in the system. From specific heat measurements, we obtained the parameters of the normal state; the Sommerfeld coefficient, $\gamma$, the electronic density of states at the Fermi level, $N(E_F)$ and the Debye temperature, $\theta_D$. In the superconducting state, the obtained values were; $\Delta C/\gamma T_c$, the electron phonon coupling, $\lambda_{e-\text{ph}}$, the superconducting energy gap, $2\Delta_0$, and the ratio $2\Delta_0/k_B T_c$. The electronic specific heat is well described by the BCS theory, suggesting that the energy gap is isotropic and in a strong coupling state.

Keywords Superconductivity · Noncentrosymmetric · Specific heat

1 Introduction

Since the discovery of superconductivity in 1911, the search for novel superconductors specially materials with higher critical temperatures have been carried out intensively. Superconducting materials can be classified in many categories: metal-based system, copper-oxygen, and iron pnictides/chalcogenides (iron-based superconductors), and others [1]. For instances, iron-based superconductors are ideal candidates for several applications of superconductivity: wires, tapes, and coated conductors for high magnetic fields. The polycrystalline compound FeSe is an interesting material for bulk applications like superconducting trapped-field magnets or super-magnets [2–4].

Noncentrosymmetric heavy fermion superconductors such as CeRhSi$_3$ [12], CeIrSi$_3$ [13], UIr [14], CePt$_3$Si [15, 16] have attracted attention because the unconventional behavior in these strongly correlated electronic compounds. On the other sides, transition-metal compounds like Li$_2$Pd$_3$B [17], Li$_2$Pd$_{3-x}$Cu$_x$B with $x = 0.0$, $0.1$ and $0.3$ [18], Li$_2$Pt$_3$B [19], Mg$_{10}$Ir$_{19}$Bi$_{16}$ [20], Rh$_2$Ga$_4$, Ir$_2$Ga$_4$ [21], Nb$_{0.18}$Re$_{0.82}$ [22] are more suitable for exploring the issue of inversion symmetry breaking. In previous reports, the ternary metallic borides Li$_2$Pd$_3$B were classified as noncentrosymmetric conventional superconductor without strong electronic correlation [17, 23, 24].

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One antecedent different to this issue is that the well-known ternary metallic boride Li2Pd3B, discovered by Togano with transition temperature about 8 K [17], was classified as noncentrosymmetric conventional superconductor without strong electronic correlation [23, 24].

In this study, we present the effect of substitution of Pd with a nonmagnetic element, in an early reported [18]. We choose the substitution of Pd with Ag, a non-magnetic element. The reason was because is isostructural and the atomic ratio of Ag is bigger than Pd. The difference in the atomic ratio should generate an internal pressure effect in the unit cell and modify the superconducting properties. In this work, we study the characteristics of this compound: crystalline structure, magnetic and electronic specific heat Li2Pd3−xAgxB with three Ag compositions; x = 0.0, 0.1 and 0.3. We compare and observe the Pauli limit with the upper critical field because is a criterion related to the importance of the spin-triplet pairing component. We estimate the parameters of the normal and superconducting state, and the strength of the electron-phonon coupling.

2 Experimental details

Polycrystalline samples of Li2Pd3−xAgxB with three compositions; x = 0.0, 0.1 and 0.3 were synthesized by two steps in an arc-melting apparatus in order to minimize losses of Li by evaporation. Stoichiometric samples were prepared using 20% of Li (99 +%), Pd (99.95%), Ag (99.95%) and B (99.99%) powders as precursors. The melting was performed in a chamber with high pure argon atmosphere. X-ray diffraction patterns were measured at room temperature in a Siemens (D5000) diffractometer with Cu Kα radiation (λ = 1.79026 Å) and Fe filter in steps of .015° at 8 s in the 2θ range of 20−110°. Rietveld analysis of the diffraction patterns was performed with MAUD program [25].

Electrical resistance versus temperature R(T) and magnetic field was measured in a Physical Properties Measurement System, PPMS (Quantum Design). R(T) without magnetic field was measured from 300 to 2 K. The magnetoresistance measurements were determined between 2 and 10 K with applied magnetic field between 0 and 40 kOe. Magnetization measurements were determined in a MPMS magnetometer (Quantum Design). Temperature dependence of the magnetic susceptibilities was investigated under zero-field cooling (ZFC) and field cooling (FC) modes.

Specific heat measurements were determined using a relaxation method between room temperature and 2 K in the PPMS system. The sample was attached to the measuring stage using Apiezon N grease to ensure a good thermal contact. The contribution to the heat capacity of the sample holder and grease was subtracted from the sample measurements.

3 Results and discussion

3.1 Structural characterization

X-ray diffraction patterns for the polycrystalline samples Li2Pd3−xAgxB with the three compositions: x = 0.0, 0.1 and 0.3 are shown in Fig. 1. The main features of the patterns correspond to a cubic structure P4332. A small amount of impurities Pd2B5 (±) and PdB2 (++) were detected. It is worth mentioning that PdB2 is non-superconducting [26].

The X-ray diffraction patterns were Rietveld-fitted considering the possibility that Ag ions can occupy Pd sites. Figure 2 shows the refined pattern of Li2Pd2.9Ag0.1B.
Superconductivity in the noncentrosymmetric Li$_2$Pd$_{3-x}$Ag$_x$B with $x = 0.0, 0.1$ and $0.3$

of RRR obtained in this study could suggest that the normal-state resistivity has a weak temperature dependence on the electrical transport and very sensitive to impurities and disorder.

The inset of Fig. 4 shows the normalized resistance with resistance value at 8 K. The superconducting transitions are sharp with a transition width of 0.21 K for the samples under study. The critical temperature was determined in the point at which the first derivative of the R(T) curve reaches its maximum value. The $T_c$ values are 7.51 K, 7.19 K and 6.99 K for samples with $x = 0.0, 0.1$ and $0.3$, respectively. It can be seen that $T_c$ decreases as the Ag content increases.

The resistance as function of temperature and magnetic field for the three studied samples in the 2–8 K temperature range under 0–4 T magnetic field is shown in Fig. 5. The transition to the superconducting state is shifting to lower

Table 1 Structural parameters obtained from the Rietveld fitting of the X-ray diffraction patterns of the studied compound with Ag at 295 K

|        | $x = 0.0$ | $x = 0.1$ | $x = 0.3$ |
|--------|-----------|-----------|-----------|
| $a$ (Å) | 6.7427(3) | 6.7548(3) | 6.7637(9) |
| $V$ (Å$^3$) | 306.6 | 308.2 | 309.4 |
| $R_e$(%) | 15.82 | 16.28 | 14.34 |
| $R_b$(%) | 12.28 | 12.72 | 11.08 |
| $R_{exp}$(%) | 10.87 | 11.98 | 10.37 |
| $\chi^2$ (%) | 1.45 | 1.35 | 1.38 |
| Li$_2$Pd$_{3-x}$Ag$_x$B (%) | 90.5590 | 97.5043 | 80.1257 |
| B$_2$Pd$_3$ (%) | 9.4490 | 2.4956 | 19.0506 |
| Pd$_2$B (%) | - | - | 0.8235 |

![Fig. 3 Magnetization as a function of temperature of Li$_2$Pd$_{3-x}$Ag$_x$B at constant magnetic field of 20 Oe](image)

![Fig. 4 Resistance versus temperature of Li$_2$Pd$_3$B, the residual resistance ratio (RRR) of this sample is 1.5. The inset shows the normalized resistance R/R(8K) curves above 2 K and up to 8 K, close to the superconductor transition, of the measured samples](image)
temperatures under the increase of DC magnetic field. From this curves, we extracted the variation of the $T_c$ with the applied field shown in Figure 6. The extrapolation of the linear fit of data the upper critical field at $T = 0$ K, $H_{c2}^{\text{linear}}(0)$, with values of $6.0 \pm 0.3, 5.2 \pm 0.1$ and $5.1 \pm 0.1$ for the three samples.

The upper critical field at $0$ K was estimated using the Werthamer–Halfand–Hohenberg theory [27]:

$$H_{c2}^{\text{WHH}}(0) = 0.693 T_c \left( \frac{dH_{c2}}{dT} \right)_{T=T_c},$$

(1)

this theory has been employed in several studies of noncentrosymmetric superconducting systems [18–20, 22, 23, 28].

The values of $\mu_0 H_{c2}^{\text{WHH}}(0)$ obtained are 4.95 T, 4.43 T and 4.29 T for $x = 0.0, 0.1$ and 0.3, respectively. The value of $\mu_0 H_{c2}^{\text{WHH}}(0)$ (4.95 T) for the sample Li$_2$Pd$_3$B is comparable with the reported value of 4.8 T [17]. From the $\mu_0 H_{c2}^{\text{WHH}}(0)$ values, we calculate the coherence length, $\xi(0)$, using the Ginzburg–Landau equation:

$$H_{c2}(0) = \frac{\Phi_0}{2\pi \xi_0^2},$$

(2)

where $\Phi_0 = 2.0678 \times 10^{-15}$ T m$^2$ is the magnetic flux quantum. The values of $\xi_0$ are 8.17, 8.64 and 8.78 nm for $x = 0.0, 0.1$ and 0.3, respectively.

The obtained Pauli limit field, $\mu_0 H_{\text{Pauli}} = \Delta_0/\mu_B \sqrt{2}$ [29], takes values of 17.83T($x = 0.0$), 16.91T($x = 0.1$) and 17.77T($x = 0.3$). It is noteworthy that these values are higher than the values obtained from the upper critical fields. Accordingly, if the upper critical field in a single crystal exceeds the Pauli limit this suggests and implies a substantial contribution from the spin-triplet component to the pairing amplitude, presumably because broken inversion symmetry [22]. However, in this study, the Pauli limiting field is greater than the upper critical fields $\mu_0 H_{c2}(0)^{\text{WHH}}$ and $\mu_0 H_{c2}(0)^{\text{linear}}$, suggesting that the Cooper pairs are in spin-singlet state. The upper critical fields values obtained from a linear extrapolation and the WHH theory show a decrease while increasing Ag substitution: As a result, a increase in $\xi_0$ values is obtained for the samples in study.

3.3 Specific heat

In the normal state, above the transition temperature, the specific heat data are well fitted by a sum of the electronic and lattice contribution [30]:

$$C_p = \gamma T + \beta T^3.$$  

(3)

A linear fit to $C_p/T$ versus $T^2$ plot of the Li$_2$Pd$_{1-x}$Ag$_x$B with $x = 0.0, 0.1$ and 0.3 is shown in Fig. 7 yielding the Sommerfeld coefficient $\gamma$ and the Debye constant $\beta$. For the sample without Ag, the value of $\gamma$ is 1.26 mJ/mol K$^2$, this value is a little bit higher than the reported previously, which varies from 8.3 to 9.8 mJ/mol K$^2$ [31, 32]. From the value of $\beta$, we estimate the Debye temperature using the relation

$$\theta_D = \left( \frac{12n\pi^4 R}{5\beta} \right)^{\frac{1}{3}},$$

(4)

where $R$ is the gas constant.
Fig. 7 $C_p/T$ versus $T^2$ curves of Li$_2$Pd$_{1-x}$Ag$_x$B: a $x = 0.0$, b $x = 0.1$ and c $x = 0.3$. The solid line represents a linear fit to the specific heat in the normal state. The inset shows $C_p/\Delta T$ versus $T$ to determine the specific heat jump at the superconducting transition.

Table 2 Parameters obtained from the specific heat of the system Li$_2$Pd$_{1-x}$Ag$_x$B ($x = 0.0, 0.1$ and $0.3$); Sommerfeld coefficient $\gamma$, density of states at Fermi level $N(E_F)$, Debye constant $\beta$, Debye temperature $\theta_D$, transition temperature $T_C$, specific heat jump $\Delta C$, electron-phonon coupling constant $\lambda_{e-ph}$, superconducting energy gap $\Delta(0)$ and the ratio $\Delta/k_B T_C$.

| $x$   | 0.0  | 0.1  | 0.3  |
|-------|------|------|------|
| $\gamma$ (mJ/molK$^2$) | 11.26 ± 0.31 | 8.11 ± 0.37 | 17.48 ± 0.26 |
| $N(E_F)$ (eV$^{-1}$) | 2.43 ± 0.065 | 1.72 ± 0.078 | 3.70 ± 0.055 |
| $\beta$ (mol/molK, K$^2$) | 1.3 ± 0.004 | 1.22 ± 0.005 | 1.38 ± 0.004 |
| $\theta_D$ (K) | 208 | 212 | 204 |
| $T_C$ (K) | 37.6 | 6.89 | 6.72 |
| $\Delta C/\gamma T_C$ | 0.96 ± 0.03 | 1.14 ± 0.05 | 0.89 ± 0.01 |
| $\lambda_{e-ph}$ | 0.84 | 0.81 | 0.82 |
| $2\Delta(0)$ (meV) | 2.92 ± 0.03 | 2.77 ± 0.02 | 2.91 ± 0.04 |
| $2\Delta(0)/k_B T_C$ | 4.60 ± 0.03 | 4.67 ± 0.03 | 5.03 ± 0.07 |

where $n$ is the number of atoms per formula unit. The value of $\theta_D$ for the Li$_2$Pd$_{1.7}$Ag$_{0.3}$ sample is 207.8 K, which agrees with the reported values of 221 K [31] and 202 K [33]. On the other hands, using the experimental Sommerfeld constant $\gamma$, the electronic density of states at Fermi level $N(E_F)$ can be deduced using the formula:

$$N(E_F) = \frac{3\gamma}{\pi^2 k_B^2 N_A}.$$  \hspace{1cm} (5)

$k_B$ is the Boltzmann’s constant and $N_A$ the Avogadro’s number. The parameters obtained are shown in Table 2. It can be seen that there is a non-monotonic variation of $N(E_F)$ and $\gamma$ as the concentration of Ag is increased. This may be due to the impurity of B$_2$Pd$_3$ on the sample Li$_2$Pd$_{2.7}$Ag$_{0.3}$B.

In the inset of Fig. 7, $C_p/T$ versus $T$ is plotted detail of the specific heat jump at the thermodynamic transition.

From the critical temperature $T_c$ and Debye temperature $\theta_D$, we can estimate the electron-phonon coupling constant $\lambda_{e-ph}$ with the Mc Millan’s relation [34] given below

$$\lambda_{e-ph} = \frac{1.04 + \mu^* \ln(\theta_D/1.45 T_c)}{(1 - 0.62 \mu^*) \ln(\theta_D/1.45 T_c) - 1.04}$$  \hspace{1cm} (6)

where $\mu^*$ represents the screened repulsive Coulomb potential, usually in the range 0.1–0.15, in this work, we used the value of 0.13. This value is normally used for intermetallic superconductors [31, 34]. The values of $\lambda_{e-ph}$ obtained in this study range from 0.84 and 0.81 for different contents of Ag. The value reported for the Li$_2$Pd$_{1.7}$B compound is $\lambda_{e-ph} = 1.09$ [31], which is higher that the obtained in this work. The values of $\lambda_{e-ph}$ are shown in Table 2 and allows to classify the system Li$_2$Pd$_{1-x}$Ag$_x$B with $x = 0.0, 0.1$ and $0.3$ in a moderated coupled superconductor state.
At low temperatures, the specific heat fit well to an exponential decay [35]:
\[ C_p = C_0 + A \exp \left( \frac{-\Delta_0}{kB T} \right), \] (7)
where \( C_0 \) is assumed as background of \( C_p \). In order to determine the superconducting energy gap, \( 2\Delta_0 \), the \( C_p(T) \) data were fitted with equation 7. The fit is shown as a continuous line in Fig. 8. The obtained values of the superconducting gap, \( 2\Delta_0 \) and \( 2\Delta_0/k_B T_c \) for the samples under study are shown in Table 2.

Several values of the superconducting energy gap of \( \text{Li}_2\text{Pd}_3\text{B} \) have been reported, from 2.55 to 3.2 meV [31, 32, 36], the value of \( 2\Delta_0 = 2.92 \) meV, obtained in this work, agrees with these values. The values of \( 2\Delta_0/k_B T_c \) obtained in this work increase from 4.60 for \( x = 0.0 \) to 5.03 for \( x = 0.3 \), these values are higher than the value predicted by BCS, \( 2\Delta_0/k_B T_c = 3.52 \), and to the reported value of 3.94 [31]. Our results indicate that the system \( \text{Li}_2\text{Pd}_{1-x}\text{Ag}_x\text{B} \) with \( x = 0.0, 0.1 \) and 0.3 is a superconducting system with strong electron-phonon coupling.

The specific heat jump at \( T_c \), \( \Delta C/\gamma T_c \) determined from \( C_p(T) \) data is lower than the BCS predicted value, 1.43, and to the reported values in \( \text{Li}_2\text{Pd}_3\text{B} \) [31, 32, 36] that varies from 1.6 to 2. Differences of \( \Delta C/\gamma T_c \) between different reports may be due to the difficulties in achieving the stoichiometry of Li and disorder in the samples.

We compare the results obtained in this work with the other published \( \text{Li}_2\text{Pd}_{1-x}\text{Cu}_x\text{B} \) with \( x = 0.0, 0.1 \) and 0.2, where the lattice parameter decreases, \( a \), as the nominal concentration of Cu increases. It is clear that the substitution of Pd with Ag and Cu generates an internal pressure in the unit cell and therefore a decrement of \( T_c \), which can be related with a decrease in the \( 2\Delta(0) \).

4 Conclusions

In this work, we reported the synthesis of three polycrystalline samples of \( \text{Li}_2\text{Pd}_{1-x}\text{Ag}_x\text{B} \) with compositions: \( x = 0.0, 0.1 \) and 0.3 prepared by arc melting technique. Rietveld analysis of the X-ray diffraction patterns shows an increase in lattice parameters as Ag content increases. Magnetization, electrical transport and specific heat measurements confirm that the superconducting transition temperature, \( T_c \), decreases as the content of Ag is increased. The upper critical fields \( \mu_0H_{c2}^{\text{WHH}}(0) \) and \( \mu_0H_{c2}^{\text{linear}}(0) \) are lower than the Pauli limiting field which suggests that the pairing is in a singlet state. The behavior of the low-temperature specific heat allows to classify this system as a superconductors with an isotropic energy gap as a conventional BCS superconductor, where the ratio, \( 2\Delta_0/k_B T_c \), values are indicative of strong coupling, contrary to the determined in other studies already published. We noted that Ag substitution decreases the electron \( N(E_F) \), observed in the decreasing of the energy gap. For the two compounds with Ag, the evidence is not so clear because we found small impurities. However, in the most pure sample, the effect is quite clear.

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Declarations

Conflict of interest The authors declare that they have not conflict of interest.

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