Superconductivity at 5.4 K in β-Bi2Pd

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Superconductivity at 5.4 K in β-Bi_2Pd

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We investigate bulk superconductivity in a high-quality single crystal of Bi_2Pd (β-Bi_2Pd; space group: $I4/mmm$) with a superconducting transition temperature of 5.4 K, by exploring its electrical resistivity, magnetic susceptibility, and specific heat. The temperature dependence of the electrical resistivity shows convex-upward behavior at temperatures greater than 40–50 K, which can be explained using a parallel-resistor model. In addition, the temperature dependences of the upper critical magnetic field and the specific heat suggest that β-Bi_2Pd is a multiple-band/multiple-gap superconductor.

KEYWORDS: superconductivity, multiple superconducting gaps, β-Bi_2Pd, Pd-Bi alloys, electrical resistivity, parallel-resistor model, upper critical field, specific heat

Studies of alloy superconductors (SCs) were of considerable interest in the 1950s and 1960s. Matthias *et al*. established the empirical law that the superconducting transition temperature, T_c, depends on the number of valence electrons; this law is widely known as the Matthias rule.[1] Among the Pd-Bi alloys, several superconducting materials, which were summarized in the review paper reported by Matthias *et al.*,[1] have been identified: α-BiPd (monoclinic structure, space group $P2_1$) with a T_c of 3.8 K; α-Bi_2Pd (monoclinic structure, space group $C2/m$) with a T_c of 1.73 K; β-Bi_2Pd (tetragonal structure, space group $I4/mmm$) with a T_c of 4.25 K;[2] and γ-phase $Pd_{2.5}Bi_{1.5}$ (hexagonal structure, space group $P63/mmc$) with a T_c of 3.7–4 K.[3] Among these alloys, the α-BiPd phase has recently been investigated as a non-centrosymmetric SC.[4] The results of studies have shown that the anisotropy of α-BiPd is not so large and that the overall effect of the no-inversion symmetry is of minor importance with respect to the bulk properties in α-BiPd.[4] However, no detailed reports concerning the physical properties of the other Pd-Bi superconducting phases, other than those that have detailed their T_c values and lattice parameters, have been published.

In this letter, we focus on one of the Pd-Bi alloys, β-Bi_2Pd, the crystal structure of which is shown in Fig. 1(a), and report the results of our investigations of a β-Bi_2Pd single crystal. An early study[2] revealed that this compound showed superconductivity at temperatures less than 4.25 K. However, we found that, by improving the crystal quality, the T_c of β-Bi_2Pd can reach 5.4 K. In addition, the temperature dependences of the upper critical magnetic field and the specific heat suggest that β-Bi_2Pd is a multiple-band/multiple-gap SC. While multigap superconductivity, where the gaps on different parts of the Fermi surface become different magnitudes, was proposed theoretically,[5] the first experimental observation of the possible existence of two distinct superconducting gaps was in the tunneling measurement of Nb-doped $SrTiO_3$.[6] The existence of multiple supercoducting gaps leads to the anomalous temperature dependences of characteristics such as the specific heat, the upper critical magnetic field, and the penetration depth.[7–13] After the discovery of the typical multigap SC, MgB_2,[7,14] numerous studies on multigap superconductivity were carried out. It is now well known that there are several multigap SCs such as $NbSe_2$,[15] $Lu_2Fe_3Si_5$,[8,11] and the iron-based SCs.[12,16] One of the interesting aspects of multigap SCs is the variety of pairing mechanisms. In iron-based SCs, the novel $s_\pm$-state, where a sign reversal of the gap function occurs between the hole and the electron pockets, has been proposed as a possible scenario.[17,18] We demonstrate that β-Bi_2Pd is also a new candidate for a multigap SC, referring to the results of the specific heat and the upper critical magnetic field.

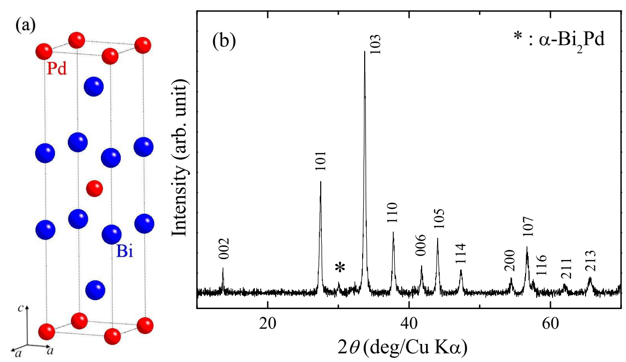

Fig. 1. (color online) (a) Schematic crystal structure of β-Bi_2Pd. (b) Powder X-ray diffraction pattern at room temperature using Cu Kα radiation for β-Bi_2Pd single crystal.

Bi_2Pd single crystals were grown via a melt-growth method. The starting materials were Bi grains (5N) and a Pd wire (3N). These materials, in the prescribed molar ratio of Bi:Pd = 2:1 (total: 2 g), were sealed in an evacuated quartz tube. This quartz tube was heated at 900

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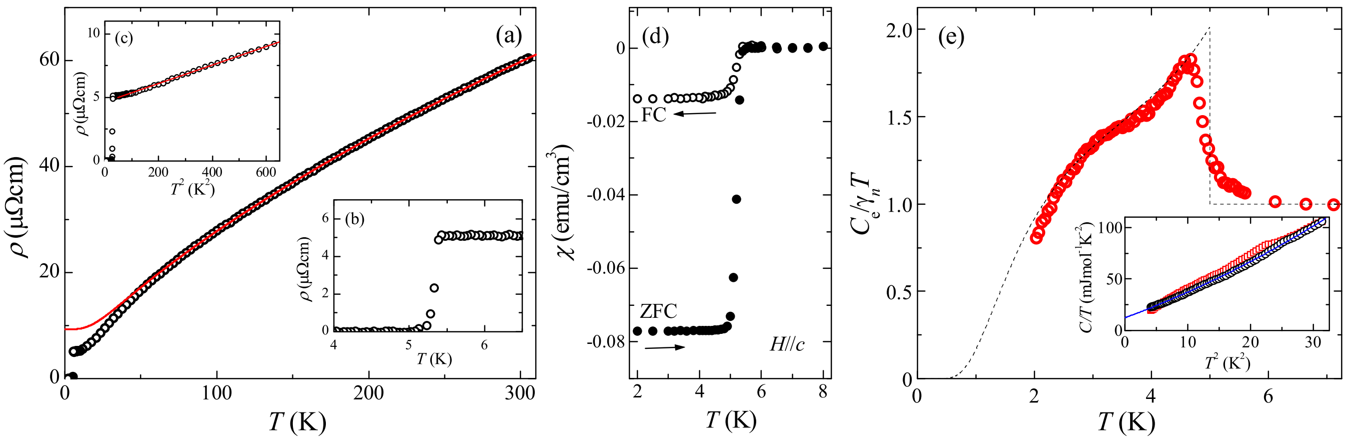

Fig. 2. (color online) (a) Temperature dependence of the electrical resistivity (ρ) of a β-Bi_2Pd single crystal. Inset (b) shows ρ near T_c; ρ at temperatures less than 25 K is plotted in inset (c) as a function of T^2. (d) Temperature dependence of the magnetic susceptibility of a β-Bi_2Pd single crystal measured in a magnetic field of 2 Oe. Closed and open circles represent the measurements in the zero-field cooling (ZFC) and field-cooling (FC) states, respectively. (e) Temperature dependence of normalized electronic specific heat in zero field. The dashed curve is calculated using the two-band model ($2\Delta_1/k_BT_c = 2.5$, $2\Delta_2/k_BT_c = 6$, $\gamma_1/\gamma_n = 0.90$), the details of which are described in the text. The specific heat divided by temperature at $\mu_0 H = 0$ T (red squares) and 0.6 T (black circles) is plotted in the inset as a function of T^2. The details of the blue solid curve are described in the text.

°C for 24 h, successively cooled to 600 °C for 72 h, and then quenched in cold water. All of the products were characterized by powder X-ray diffraction (XRD) using Cu Kα radiation at room temperature. Magnetic susceptibility measurements were performed using a superconducting quantum interference device (SQUID) magnetometer. The electrical resistivity, ρ, was measured by the four-terminal method over the temperature range of 0.5 to 300 K under magnetic fields as strong as 3 T. The specific heat was measured by the thermal-relaxation method at temperatures as low as 2 K on a commercial apparatus (Physical Property Measurement System, Quantum Design).

Figure 1(b) shows the XRD pattern of a Bi_2Pd single crystal. Except for a few peaks that resulted from α-Bi_2Pd, all of the peaks were indexed on the basis of a tetragonal lattice (no. 144, $I4/mmm$) with $a = 3.37$ Å and $c = 12.96$ Å. These lattice parameters are in good agreement with those reported previously.[2,19]

The temperature dependence of ρ for a β-Bi_2Pd single crystal is shown in Fig. 2(a). The large residual resistivity ratio [RRR, $\rho(T = 300\text{ K})/\rho(T = 6\text{ K})$] of 12 indicates the high quality of the crystal. The T_c^{onset}, which is defined as the temperature at which ρ begins to deviate from the normal-state behavior, and the T_c^{zero}, which is defined as the temperature at which ρ becomes zero, were estimated to be 5.4 and 5.3 K, respectively, as shown in Fig. 2(b). These values are greater than the value of 4.25 K reported in previous papers.[2,19] The temperature dependence of the magnetic susceptibility in a magnetic field of 2 Oe is shown in Fig. 2(d). This result reveals that the diamagnetic transition of β-Bi_2Pd occurs at a temperature less than 5.4 K, which is in good agreement with the $\rho(T)$ data. Here, it is interesting to note that the T_c of β-Bi_2Pd reported here is almost the same as that of Pd-intercalated Bi_2Te_3 with a very small superconducting volume fraction ($< 1\%$) in ref. 20, where the possibility that the topological insulator Bi_2Te_3 can be made into an SC by Pd intercalation between the Bi_2Te_3 layers is argued.

The temperature dependence of ρ for β-Bi_2Pd exhibits the convex-upward characteristics at temperatures greater than 50 K; these characteristics are similar to those observed for A15 SCs.[21–24] Fisk and Webb have proposed that the resistivity of A15 compounds at high temperatures saturates at a value, ρ_{sat}, that corresponds to the mean free path on the order of the interatomic spacing.[23] Wiesmann *et al.*[25] developed the idea proposed by Fisk and Webb and found empirically that the ρ of A15 compounds could be described using a parallel-resistor model:

$$\rho\left(T\right) = \left[\frac{1}{\rho_{\text{sat}}} + \frac{1}{\rho_{\text{ideal}}\left(T\right)}\right]^{-1}, \tag{1}$$

where ρ_{sat} is the resistivity saturated at high temperature and is independent of T, and $\rho_{\text{ideal}}(T)$ is the "ideal" contribution according to Matthiessen's rule, $\rho_{\text{ideal}}(T) = \rho_{\text{ideal},0} + \rho_{\text{ideal,L}}(T)$. Here, $\rho_{\text{ideal},0}$ is the ideal temperature-independent residual resistivity caused by impurity scattering. $\rho_{\text{ideal,L}}(T)$ is the temperature-dependent contribution caused by thermally excited phonons and can be expressed by the Bloch-Grüneisen formula or by Wilson's theory:[26,27]

$$\rho_{\text{ideal,L}}\left(T\right) = C_1\left(\frac{T}{\theta_D}\right)^r \int_0^{\frac{\theta_D}{T}} \frac{x^r}{\left(e^x - 1\right)\left(1 - e^{-x}\right)}dx, \tag{2}$$

where C_1 is a numerical constant, θ_D is the Debye temperature, and the values of the exponent r are 3 and 5 for Wilson's theory and the Bloch-Grüneisen formula, respectively. The data for ρ from 300 to 75 K were fitted to eq. (1), and the fitted result is shown in Fig. 2(a) as the solid curve. For $\rho_{\text{ideal,L}}(T)$, we found that a better fit for $\rho(T)$ in β-Bi_2Pd is given by Wilson's expression [specifically, $r = 3$ in eq. (2)], which takes into account the interband electron-phonon Umklapp scattering between a low-mass s-band and a heavy-mass d-band.[27] The best-fitted result yields the values of 134 K for θ_{D}, 241 $\mu\Omega$cm for ρ_{sat}, 9.63 $\mu\Omega$cm for $\rho_{\mathrm{ideal},0}$, and 63.3 $\mu\Omega\mathrm{K}^{-3}$ for C_1. The value of θ_{D} is very close to that obtained from the specific heat measurement, as will be discussed later. These results show that the parallel-resistor model explains the $\rho\,(T)$ behavior of β-$\mathrm{Bi_2Pd}$ well at high temperatures. In contrast, notable deviations between the experimental data and the parallel-resistor model are observed at low temperatures. In Fig. 2(c), ρ is plotted as a function of T^2 at low temperatures, which shows that the resistivity is proportional to T^2 at temperatures less than 25 K. A similar crossover from the T^2 behavior to the saturated behavior upon heating has been observed in A15 compounds such as $\mathrm{Nb_3Sn}$[21, 28] and in β-pyrochlore oxides, $A\mathrm{Os_2O_6}$ (A =K, Rb, Cs).[29] Some mechanisms of the T^2-dependence of $\rho\,(T)$ have been proposed.[30–35] However, the origin of the T^2-dependence of ρ in β-$\mathrm{Bi_2Pd}$ cannot be specified solely from the results presented in this letter; further studies are needed.

Next, the specific heat divided by temperature, C/T, at $\mu_0 H = 0$ (red squares) and 0.6 T (black circles) is plotted in the inset of Fig. 2(e) as a function of T^2. C/T at $\mu_0 H = 0.6$ T, where superconductivity is fully suppressed above 2 K, was fitted to the expression

$$C = \gamma_n T + \beta_n T^3 + \alpha_n T^5, \tag{3}$$

where $\gamma_n T$ is the electronic term, C_{e}, and $\beta_n T^3 + \alpha_n T^5$ represents the phonon contribution. From the fitting with eq. (3), which is shown in the inset of Fig. 2(e) as the blue solid curve, we obtained the parameters $\gamma_n = 12$ $\mathrm{mJmol^{-1}K^{-2}}$, $\beta_n = 2.3$ $\mathrm{mJmol^{-1}K^{-4}}$, and $\alpha_n = 0.02$ $\mathrm{mJmol^{-1}K^{-6}}$. The existence of the T^5 term in the normal-state specific heat suggests a complex phonon density of states. From this value of β_n, θ_{D} was estimated to be 136 K using the relation $\theta_{\mathrm{D}} = \left(12\pi^4 N k_{\mathrm{B}}/5\beta_n\right)^{1/3}$,[26] where N is the number of atoms, and k_{B} is the Boltzmann constant. This value of θ_{D} is similar to that obtained from the analysis of the $\rho(T)$ data using eq. (1), as previously mentioned. The temperature dependence of normalized electronic specific heat at $\mu_0 H = 0$ T, which is estimated using the above parameters, is shown in Fig. 2(e). A clear jump appeared in $C_{\mathrm{e}}/\gamma T$ at a temperature of 5.0 K. This value is slightly lower than the T_{c} estimated from the temperature dependences of ρ and χ. The magnitude of the jump at $T = T_{\mathrm{c}}$, ΔC, is 40 $\mathrm{mJmol^{-1}K^{-1}}$, and the value of the normalized specific-heat jump, $\Delta C/\gamma_n T_{\mathrm{c}}$, is 0.82. This value is smaller than that expected in the simple BCS weak-coupling limit, i.e., 1.43. In addition, C_{e} of β-$\mathrm{Bi_2Pd}$ below T_{c} shows a peculiar temperature dependence. That is, there is a plateau at approximately 3 K. One might conclude that this plateau results from some impurity phases, for example, amorphous Bi or Bi-Pd alloys other than β-$\mathrm{Bi_2Pd}$. However, there is no anomaly in $\chi(T)$ at $T \sim 3$ K. Thus, it is unlikely that the origin of this plateau in the normalized electronic specific heat is an impurity phase. These features, that is, the small jump at T_{c} and the plateau at approximately 3 K, in $C(T)$ of β-$\mathrm{Bi_2Pd}$ are familiar in the multigap SCs.[7, 8, 36] In the case of an SC with a single gap, the entropy, S, and C are described as follows:[37]

$$\frac{S}{\gamma_n T_{\mathrm{c}}} = -\frac{6}{\pi^2 k_{\mathrm{B}} T_{\mathrm{c}}} \int_0^\infty \left[f \ln f + (1-f)\ln(1-f)\right] d\epsilon, \tag{4}$$

$$\frac{C}{\gamma_n T_{\mathrm{c}}} = t\frac{d\left(S/\gamma_n T_{\mathrm{c}}\right)}{dt}, \tag{5}$$

where $f = \left[\exp\left(E/k_{\mathrm{B}}T\right) + 1\right]^{-1}$. The energy of quasi particles is given by $E = \left[\epsilon^2 + \Delta^2\,(t)\right]^{0.5}$, where ϵ is the energy of the normal electrons relative to the Fermi surface and $\Delta\,(t) = \Delta_0 \delta\,(t)$ is the temperature dependence of the gap energy. Here, $\delta\,(t)$ is the normalized BCS gap at the reduced temperature, $t = T/T_{\mathrm{c}}$.[38] For the analysis of the data for β-$\mathrm{Bi_2Pd}$, we use the two-band, two-gap model, where the total specific heat is considered as the sum of the contributions of each band calculated independently according to eq. (5), as in the cases of $\mathrm{MgB_2}$ and $\mathrm{Lu_2Fe_3Si_5}$.[7, 8] Each band is characterized by the Sommerfeld coefficient, γ_i, with $\gamma_1 + \gamma_2 = \gamma_n$. We calculate the specific heat by this two-gap model using three parameters of two gaps (Δ_1, Δ_2) and the relative weights ($\gamma_1/\gamma_n \equiv x$, $\gamma_2/\gamma_n \equiv 1-x$), and one of the calculated results is shown as the dashed curve in Fig. 2(e). The curve calculated using the two-gap model is in agreement with the experimental data, at least above 2 K, which suggests that β-$\mathrm{Bi_2Pd}$ is a multigap SC. In this analysis, however, there is still some uncertainty and it is difficult to determine only one set of three parameters for lack of experimental data of C at temperatures less than 2 K. A more detailed analysis requires data at lower temperatures, and such measurements are currently in progress.

The effect of a magnetic field on ρ is shown in Fig. 3(a). The T_{c} decreases almost linearly with increasing magnetic field. The upper critical field, $\mu_0 H_{\mathrm{c2}}$, which is defined as the field in which ρ becomes half the value of the normal-state resistance, is plotted in Fig. 3(b) as a function of temperature. The upper critical field extrapolated to $T = 0$ K, namely, $\mu_0 H_{\mathrm{c2}}(0)$, is estimated to be 1.13 ± 0.05 T($H_{\mathrm{c2}}^{ab}(0)$) and 0.73 ± 0.05 T($H_{\mathrm{c2}}^{c}(0)$) for magnetic fields parallel and perpendicular to the ab-plane, respectively. These results give Ginzburg-Landau coherence lengths of $\xi_{ab}(0) \sim 212 \pm 8$ Å and $\xi_c(0) \sim 137 \pm 2$ Å, using $\mu_0 H_{\mathrm{c2}}^{ab}(0) = \Phi_0/2\pi\xi_{ab}(0)\xi_c(0)$ and $H_{\mathrm{c2}}^{c}(0) = \Phi_0/2\pi\xi_{ab}(0)^2$, where $\Phi_0 = 2\pi\hbar/2e = 2.07 \times 10^{-15}$ $\mathrm{Tm^2}$ is the magnetic flux quantum. The anisotropy parameter, Γ, which is defined as $\Gamma = H_{\mathrm{c2}}^{ab}(0)/H_{\mathrm{c2}}^{c}(0)$, is found to be 1.6. It should be noted that the temperature dependence of $\mu_0 H_{\mathrm{c2}}$ in β-$\mathrm{Bi_2Pd}$ reveals a positive curvature close to T_{c}, which becomes negative at temperatures less than approximately 3 K, as shown in Fig. 3(b). These temperature dependences have also appeared in other multigap SCs, such as $\mathrm{MgB_2}$,[10, 39] LaFeAs(O,F),[12, 40] and SrPtAs.[41] In addition, in some theoretical papers, this temperature dependence of $\mu_0 H_{\mathrm{c2}}$ has been explained on the basis of multiple superconducting gaps.[42–44] This result

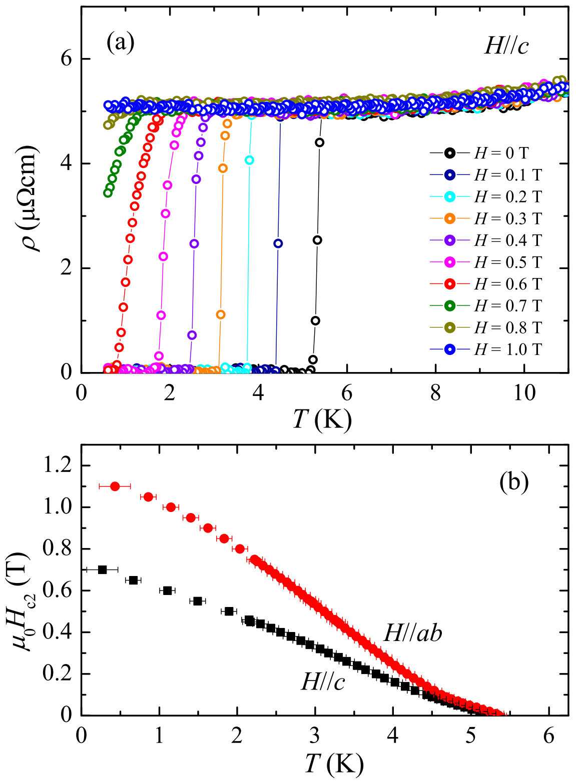

Fig. 3. (color online) (a) Temperature dependence of the electrical resistivity of a β-Bi_2Pd single crystal in a magnetic field parallel to the c-axis. The upper critical field, $\mu_0 H_{c2}$, is plotted in (b) as a function of temperature. Closed circles and squares represent the $\mu_0 H_{c2}(T)$ data for magnetic fields parallel to the ab-plane and to the c-axis, respectively.

for $\mu_0 H_{c2}$, together with the $C_e\,(T)$ data, suggests that β-Bi_2Pd is an SC with multiple superconducting gaps. Indeed, the presence of different Fermi surfaces has already been predicted by a band calculation.[45] Thus, our experimental findings suggest that the superconducting gaps open on different Fermi surfaces with different magnitudes.

In conclusion, we observed bulk superconductivity with a T_c of 5.4 K in β-Bi_2Pd by investigating the electrical resistivity, the magnetic susceptibility, and the specific heat. The value of T_c reported in this letter is higher by approximately 1.2 K than those reported in previous papers and is the highest among the Pd-Bi alloy systems. In addition, the temperature dependences of the upper critical field and the specific heat suggest that β-Bi_2Pd is a multigap superconductor.

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