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Superconductivity in the Heusler alloy Pd_2YbPb

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Abstract

Magnetization and resistivity measurements have been carried out on the Heusler alloy Pd_2YbPb in the temperature range 2–300 K. Both low-field dc susceptibility and resistivity show that this material undergoes a superconducting transition at 2.85 K. The presence of superconductivity was also confirmed by the Meissner effect observed at temperatures below 3 K. The temperature dependence of the susceptibility and resistivity have been accounted for on the basis of a crystal electric field with a Γ_7 ground state. Both measurements suggest that the energy splitting between a Γ_7 doublet and Γ_8 quartet, the first excited state, is about 65 K. However, the energy splitting between Γ_7 and Γ_6 (a doublet at highest energy) deduced from the resistivity data is 165 K, while the susceptibility data yield 355 K. We ascribed this discrepancy to the poor fitting quality of susceptibility at higher temperatures. We compare the results with those published in the literature for the isoelectronic Heusler compound Pd_2YbSn.

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1. Introduction

Ishikawa et al. [1] in 1982 first synthesized superconducting Heusler alloys containing palladium and heavy rare-earth elements for a study of triplet pairing superconductivity. At that time, the authors concluded that in these Heusler alloys, the d-electrons of the palladium atoms with a large Stoner factor are mainly responsible for the strong coupling superconductivity. They also found that for Pd_2YbSn, magnetic ordering occurs at about 0.26 K in the superconducting state. The superconducting transition temperature T_c was at a temperature above 1 K. However, the nature of the magnetic state was not discussed in detail. Following this study, Kierstead et al. [2] and Malik et al. [3] have reported the coexistence of superconductivity and magnetic ordering in zero magnetic field for the same compound. Aoki et al. [4] have reported specific heat, thermoelectric power, magnetization, and electrical transport measurements on Pd_2YbSn. The results of these measurements lead them to suggest that a weak magnetic coupling between the Pd sublattice and the Yb–Sn sublattice is present being responsible for superconductivity and antiferromagnetic ordering. Later, Amato et al. [5] have carried out muon spin relaxation and inelastic neutron scattering experiments on Pd_2YbSn. They have pointed out to the possibility of interplay between superconductivity and magnetic fluctuations in this compound.

The experimental results mentioned above have motivated us to search for new Heusler alloys. Some have already been prepared and investigated in relation to the concept of magnetically mediated superconductors. On closer examination, we find that very few Heusler alloys of the type Pd_2RPb (isoelectronic to Pd_2RSn), which are expected to exhibit this kind of superconductivity, have been studied. There is a single study in the literature [6], to the best of our knowledge, reported on the Heusler alloys Pd_2MPb (M: rare earth, Th, and U). The coexistence of ferromagnetism and superconductivity was found only in the compound Pd_2UPb, which is also expected to have heavy fermion properties. In this study, the superconducting transition for the Pd_2YbPb at $T = 2.8$ K has been already reported by magnetic susceptibility. A magnetic contribution to the magnetic susceptibility of Pd_2YbPb down to 1.8 K has been also observed. However, this magnetic contribution has been attributed

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as a magnetic background contribution arising from the paramagnetic free Yb ions.

Heusler alloys form a group of ternary intermetallic compounds with the general formula X_2YZ. These alloys crystallize in the $L2_1$ crystal structure, which is a superstructure based on the bcc lattice. The Y-atoms have X-atoms as nearest neighbors, the Z-atoms as second nearest neighbors, and other Y-atoms as third nearest neighbors. Generally, the X-atoms are the elements of the Cu, Ni, or Co columns, the Z-atoms are non-transition s–p elements such as Si, Ge, Sn, and Pb, and the Y-atoms are other transition elements. Here we consider Heusler compounds in which the Y-site is occupied by rare-earth (R) elements such as Ce, Pr, Nd, Tm, and Lu. For X = Ni or Pd (only), the R atoms are well separated from each other and are well localized. However, hybridization between the itinerant 3d (and/or 5d) electrons and the more localized 4f or 5f electrons may induce magnetic instabilities in these systems, so that magnetic fluctuations become strong; a situation designated as the quantum criticality. These are believed to play the vital role for the coexistence of ferromagnetism and superconductivity. The magnetic instability is suggested to be one of the necessary conditions.

For the reasons given above, we have found it interesting to re-examine the electrical and magnetic properties of the Heusler alloy Pd_2YbPb. We observe from magnetization and resistivity data two successive superconducting transitions at 7 K and 2.85 K. The former is related to segregated lead paths, and the latter is intrinsic to Pd_2YbPb grains bounded by the lead paths. We present evidence for the presence of superconductivity in Pd_2YbPb by means of magnetization and resistivity measurements.

2. Experimental

Polycrystalline samples were prepared by arc melting the constituents Pd:3N, Yb:3N, and Pb:4N under argon atmosphere in a water-cooled copper crucible. The resulting ingot was cut into dimension 2 mm × 8 mm × 1 mm and annealed under argon at one-third atmospheric pressure at 800 °C for 60 days. The sample was characterized by scanning electron microscopy, and microprobe analysis. The same sample was also used in the electrical resistivity and magnetization measurements. At the end of the experiments, the sample was pulverized for X-ray studies. Fig. 1 shows a micrograph of the scanning electron microscope image. Microprobe analysis on this surface reveals Pb precipitates having needle-like morphology that have segregated to the grain boundaries of the essentially stoichiometric Heusler compound $Pd_{49.2}Yb_{27.4}Pb_{23.4}$ (nominally hereafter referred to as Pd_2YbPb). The various regions are labeled in the figure. One also observes regions appearing as cracked surfaces. These are oxide compounds of Pb and Yb with oxygen content of about 70%. No trace of Pb_2Pd, which is superconducting at 3 K, has been found [6]. X-ray powder diffraction experiments were carried out using a Co target source with the sample located on a zero background silicon carrier.The lattice parameter of the Heusler phase was determined to be 6.7390 Å. The diffraction pattern is seen in Fig. 2, where the Bragg reflections of the Heusler phase are indexed. The down-arrows point to the positions of the reflections from Pb, and the up-arrows indicate oxide phases. These oxide peaks that are associated with the areas shown in the micrograph in Fig. 1. Within our present measurements, it is not possible to identify the individual oxide species. The only non-oxidic phase we observe is the Heusler. Due to the slight off-stoichiometric composition from that of the ideal Heusler composition, a certain amount of disorder can be present. However, the presence of the superstructure reflections gives evidence for the ordered $L2_1$ Heusler phase among any trace disorder.

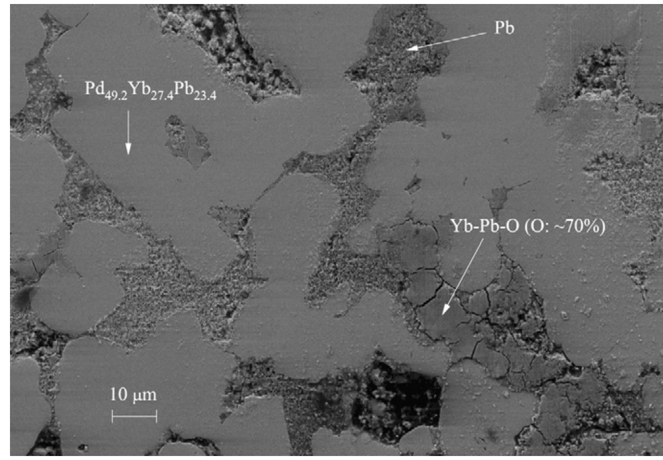

Fig. 1. Scanning electron microscope image of the sample used in the present measurements. The $Pd_{49.2}Yb_{27.4}Pb_{23.4}$ Heusler phase is surrounded by Pb paths. Oxides are also encountered.

The temperature dependence of the resistivity $\rho(T)$ was performed using conventional four probe dc technique in the temperature range of $2\,K \leq T \leq 300\,K$. The measuring current was 0.5 mA. A calibrated germanium resistance thermometer was used to measure the temperature below 80 K. For higher temperatures, a calibrated platinum resistance thermometer was used. The electrical contact was made using silver paint and 25 μm gold wire. The temperature dependence of the magne-

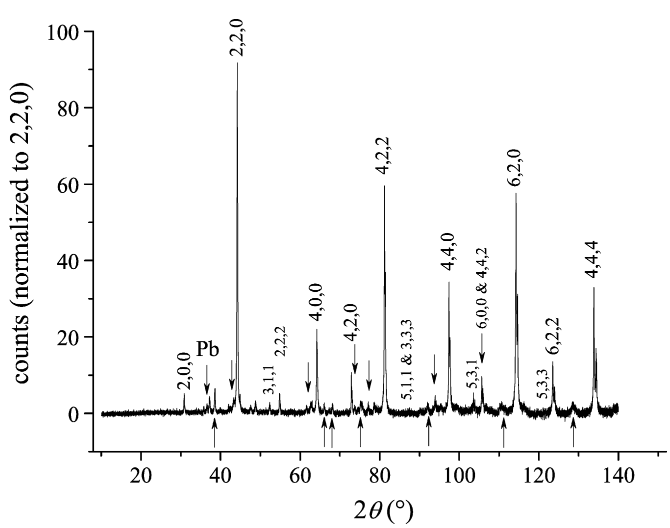

Fig. 2. The X-ray diffraction pattern of the sample. The indices refer to the Heusler phase. Reflections shown at the positions by down-arrows are those due to Pb. Up-arrows indicate the peak positions of oxides.