Journal of Alloys and Compounds 762 (2018) 254–259

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Ferromagnetism in the orthorhombic PrPd and SmPd

Vijay Dhar [a], A. Provino [b, c, *]

[a] Department of Physics, Jai Hind College, A-Road, Churchgate, Mumbai, 400 020, India
[b] Institute SPIN-CNR, Corso Perrone 24, 16152, Genova, Italy
[c] Department of Chemistry, University of Genova, Via Dodecaneso 31, 16146, Genova, Italy

ARTICLE INFO

Article history:
Received 19 July 2017
Received in revised form
8 May 2018
Accepted 16 May 2018
Available online 17 May 2018

Keywords:
PrPd and SmPd intermetallics
Rare earth alloys and compounds
Magnetic measurements
Ferromagnetism
Heat capacity
Electrical resistivity

ABSTRACT

In continuation of our studies on the binary RPd compounds (orthorhombic CrB-type structure, Pearson symbol *oS*8, space group *Cmcm*, No. 63), we report here the magnetic properties of the iso-structural PrPd and SmPd. The two compounds order ferromagnetically with the Curie temperature, T_C, of nearly 12 K and 22 K, respectively (inferred from the peak temperature of the heat capacity at the paramagnetic to ferromagnetic transition). Anomalies in the magnetization and the electrical resistivity are also observed in correspondence of the bulk magnetic transition. The ferromagnetic nature of the magnetic transition is clearly indicated by the temperature and field dependence of the magnetization. The coercive fields of the two compounds are measured to be 160 Oe and 12 kOe at 2 K, respectively. The temperature dependence of both the heat capacity and electrical resistivity indicates a gapped ferromagnetic magnon spectrum with the gaps of ≈ 11 K and ≈ 40 K in PrPd and SmPd, respectively. The data obtained in current work are compared and discussed together with those previously reported for CePd and NdPd; the Curie temperatures of the four RPd iso-structural compounds follow the *de Gennes* scaling.

1. Introduction

Although the R-Pd (R = rare earth) binary systems have been investigated for many years [1–5], their magnetic properties have not been studied extensively. Loebich and Raub investigated the R-Pd phase diagrams for R = Y, Sm, Gd, Dy, Ho and Er [1], while Iandelli and Palenzona determined those of the Eu-Pd [6] and Yb-Pd systems [7]. An updated version of the Yb-Pd phase diagram has been recently published [8]. This latter work also anticipates the existence of two new series of compounds: the R_3Pd_5 [9] and $R_{10}Pd_{21}$ [10]. Among the binary R-Pd compounds, the equiatomic RPd are reported to form for all of R (Sc, Y and all lanthanides) [3,5]. For R = La-Nd and Sm [11], Eu [6,12], Gd [13] and Tb [11] they crystallize in the orthorhombic CrB-type structure (Pearson symbol *oS*8, space group *Cmcm*, No. 63), while for heavier R, Sc and Y the structure changes to the cubic CsCl-type (*cP*2, $Pm-3m$, No. 221) [11,14,15].

Despite their simple composition, the literature data show that magnetic behavior of most RPd compounds has not been studied in detail. CePd is a Kondo compound [16]; it is shown to be a ferromagnet with a Curie temperature, T_C, of 6.5 K and undergo a second transition at 3.5 K, probably due to the reorientation of the magnetic moments [17]. In EuPd the Eu ions are divalent [18], while TbPd orders likely antiferromagnetically near 50 K [19]. Interestingly, Yb ions in YbPd are reported to be present in both Yb^{2+} and Yb^{3+} valence states [20]. We have recently investigated the magnetic behavior of NdPd; this compound orders ferromagnetically near 15 K [21]. In continuation of our studies on the RPd compounds, we report here the physical properties (magnetic susceptibility, heat capacity and electrical resistivity) of the two PrPd and SmPd compounds. Similarly to NdPd and CePd, which are both ferromagnetic, the Pr and Sm analogs also adopt a ferromagnetic ground state with Curie temperatures of 15 and 22 K, respectively.

2. Experimental

Three equiatomic RPd samples for R = La, Pr and Sm (total mass of 2.5–3.5 g) were prepared starting from high purity commercial metals (99.9 wt.% purity for R and 99.99 wt.% purity for Pd). The LaPd and PrPd alloys were synthetized by arc-melting the metals, weighed in stoichiometric proportion, under a pure TiZr-gettered Ar atmosphere. The buttons were re-melted three times after turning them upside-down to ensure homogenization; total weight loss was below 0.2 wt.%. The ingots, placed in an out-gassed Ta tube

* Corresponding author. Institute SPIN-CNR, Corso Perrone 24, 16152, Genova, Italy.
E-mail address: alessia.sting@gmail.com (A. Provino).

https://doi.org/10.1016/j.jallcom.2018.05.181
0925-8388/

and closed under vacuum in a quartz ampoule, were annealed at 700 °C for 9 days in a resistance furnace. The alloys were then removed from the furnace and air cooled. The SmPd alloy was synthetized into an out-gassed Ta crucible (sealed by arc welding under a pure Ar flux) by melting the metals, in form of small pieces, in a high-frequency induction melting furnace. This alloy was annealed at 800 °C for 10 days, then air cooled. Final bulk samples had a steel-like color and were slightly air sensitive. They also were rather compact despite their relative ductility.

The samples were checked by light optical and scanning electron microscopies (LOM and SEM); the phases analyzed by electron microprobe (EDX) for semi-quantitative analysis. EDX analyses were performed on at least three points (or areas) to identify the phases and determine their composition; the precision of the measurements was estimated to be within 0.7 at.%. Microphotographs of the samples were taken both by backscattered and secondary electrons. Structural characterization was carried out by X-ray powder diffraction (XRD) using a Guinier camera [Cu Kα1 radiation; Si as an internal standard, $a = 5.4308$ (1) Å]. It was difficult to obtain good X-ray powder patterns due to the ductility of the alloys and their sensitivity to air and moisture. The Guinier powder patterns were of quality just enough to be indexed. The patterns were indexed by the help of LAZY PULVERIX program [22] and unit cell parameters obtained by a least-squares program.

The magnetization as a function of temperature (between 1.8 K and 300 K) and magnetic field (up to a maximum of 70 kOe) was measured on a Magnetic Property Measurement System (SQUID, Quantum Design). The heat capacity (in the temperature range of 1.9 K–50 K) and electrical resistivity (between 1.9 K and 300 K) were measured on a Physical Property Measurement System (PPMS, Quantum Design).

3. Results and discussion

3.1. PrPd

The samples, prepared by following the procedure described above, resulted to be almost single phase. In some portions of the samples trace amount of R_3Pd_4 was present as a secondary phase (about 2–3 vol.%, as identified by EDX microprobe); the microstructure of the two PrPd and SmPd samples is shown in Fig. 1a and b, respectively. However, both Pr_3Pd_4 and Sm_3Pd_4 compounds have been reported not to show any magnetic ordering down to 4.2 K [23]. The lattice parameters of RPd compounds are known to vary as per the lanthanide contraction (excepting EuPd, where Eu ions are in the divalent state). The parameters obtained in this work for the three compounds are: $a = 3.948$ (2) Å, $b = 11.035$ (5) Å and $c = 4.664$ (3) Å for LaPd; $a = 3.861$ (3) Å, $b = 10.834$ (4) Å and $c = 4.609$ (2) Å for PrPd; $a = 3.781$ (2) Å, $b = 10.646$ (4) Å and $c = 4.583$ (2) Å for SmPd. They are in good agreement with values reported in literature [24]. It is therefore expected that the R ions in both PrPd and SmPd would formally be in their trivalent state. The magnetization data, indeed, confirm that at low temperatures both PrPd and SmPd compounds own magnetic ground state due to the magnetic ordering of the magnetic moments associated with Pr^{3+} and Sm^{3+} ions. The inverse magnetic susceptibility, χ^{-1}, of PrPd measured in between 1.8 K and 300 K, and in an applied magnetic field of 3 kOe is shown in Fig. 2. The inverse susceptibility of PrPd, like that of NdPd [21], shows a minor convex curvature at high temperatures, which may have the same physical origin(s) as suggested for NdPd. Namely, due to the orthorhombic symmetry, PrPd may be characterized by a strong magnetic anisotropy, resulting in a Curie-Weiss behavior of susceptibility along the easy axis and a weaker temperature dependent susceptibility along the hard direction. Overall it may lead to a gentle curvature in the χ^{-1}

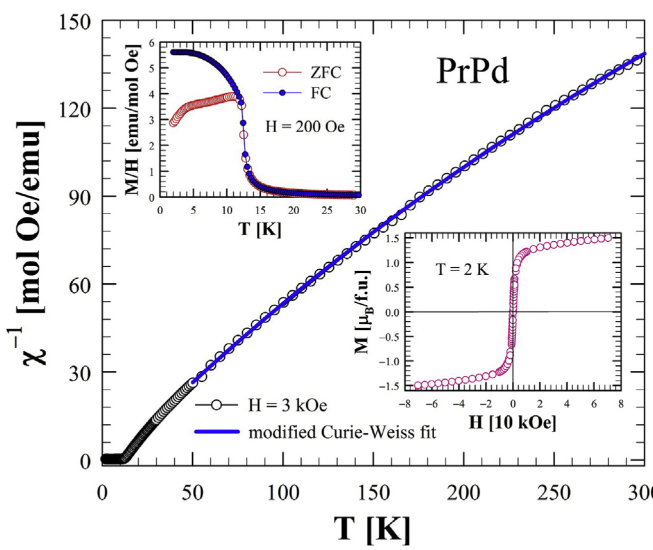

Fig. 2. The inverse magnetic susceptibility of PrPd between 1.8 K and 300 K in an applied field of 3 kOe; the blue line is the fit to the modified Curie-Weiss law. The upper inset shows the low field (200 Oe) ZFC and FC magnetization below 30 K; the lower inset shows the isothermal magnetization of PrPd at T = 2 K measured up to 70 kOe. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

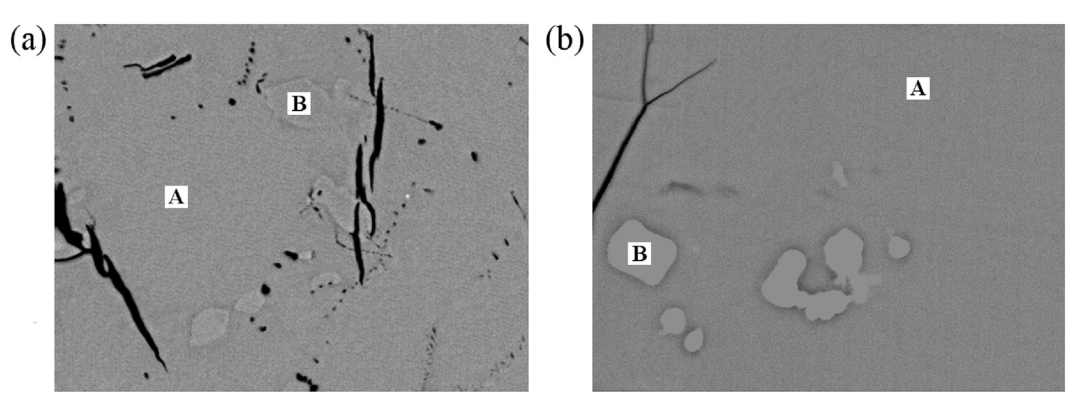

Fig. 1. SEM microphotographs [backscattered electron (BSE) mode] of the microstructure of the two PrPd (a) and SmPd (b) samples, where only the portion containing more of the extra phase is shown. In both samples the primary main phase (A) is RPd, while the lighter grains (B) are the R_3Pd_4 compound.