Crystalline Electric Field Effects in Pr\(_{1-x}\)La\(_x\)Os\(_4\)Sb\(_{12}\)

C. R. Rotundu and B. Andraka

Department of Physics, University of Florida, PO Box 118440, Gainesville, Florida 32611-8440, USA

Abstract. The specific heat of Pr\(_{1-x}\)La\(_x\)Os\(_4\)Sb\(_{12}\) alloys was measured in fields to 14 T. CEF energies remain approximately constant across this system. The field-induced AFQ order is suppressed by the alloying and is not observed for \(x=0.2\) above 0.4 K.

Keywords: CEF, heavy fermions, quadrupolar order, magnetic phase diagram

PACS: 65.40.Ba, 71.27.+a, 74.62.Dh

The discovery\([1]\) of superconductivity and heavy fermion behavior in PrOs\(_4\)Sb\(_{12}\) opens new frontiers for heavy fermion physics. A non-magnetic crystalline electric field (CEF) ground state of Pr seems to preclude a conventional mechanism leading to the formation of heavy electrons, based on a Kondo impurity model. Thus, the outstanding question is the nature of the heavy fermion state in this compound. Proximity of the low temperature state to the antiferro-quadrupolar (AFQ) order\([2]\) suggests either fluctuations of this order parameter or the quadrupolar Kondo-effect as the primary candidates. Our recent investigation\([3]\) of Pr\(_{1-x}\)La\(_x\)Os\(_4\)Sb\(_{12}\) demonstrated that heavy fermion character is strongly reduced by relatively small amounts of La substituted for Pr, despite the rather weak, if any, effect of this substitution on accessible single-ion parameters. For instance, there is an unusually small variation of the lattice constant. Similarly, the temperature of the maximum in the susceptibility (approximately 4 K) does not show any systematic variation on \(x\). This maximum is associated with excitations between the lowest CEF levels. The low temperature susceptibility hence is consistent with a CEF scheme of Pr, independent of the La content. Thus, the zero field alloying study implies a non-single impurity origin of the heavy fermion state.

The purpose of the current investigation is to determine how the field-induced AFQ order is affected by alloying and to provide further insight on how the single-ion parameters of Pr\(_{1-x}\)La\(_x\)Os\(_4\)Sb\(_{12}\) depend on \(x\). Specific heat for alloys with \(x=0.02, 0.1, 0.2,\) and 0.6 was measured in fields to 14 T.

There are compelling evidences\([4, 5]\) that the CEF ground state of PrOs\(_4\)Sb\(_{12}\) is a singlet and the first excited state is a triplet at about 8 K. Magnetic field splits the triplet and lowers its lowest energy eigenstate with respect to the singlet such that the two should cross in a field of about 9 - 10 T. Instead, when the two levels are sufficiently close in energy, the AFQ order is observed between 4 and 12 T. Thus, the long range order removes the degeneracy of a quasi-doublet in magnetic fields.

Figure 1 shows the magnetic phase diagram for \(x=0, 0.02,\) and 0.1 for fields up to 14 T. Closed and open circles represent the Schottky and AFQ anomalies, respectively, for the undoped compound. When 2 % of La is substituted for Pr, both the size of the AFQ anomaly and its temperature are reduced with respect to \(x=0\) for identical fields. For instance, the reduction of the transition temperature in fields 8 and 9.5 T is 15 - 20 % (Fig. 1). This is a modest change in comparison to PrPb\(_3\), another well studied Pr-material exhibiting AFQ order in the zero field\([6]\). In the latter case, 2 % of La is sufficient to suppress the long range order completely.

The f-electron specific heat for \(x=0.1\) is shown in Fig. 2. Broad maxima in \(H=6, 12,\) and 14 T are clearly Schottky anomalies due to excitations between the lowest CEF levels. On the other hand, peaks at 8 and 10 T are somewhat sharper and of higher magnitude, although this magnitude is considerably reduced with respect to \(x=0\) and 0.02. These peaks most probably represent some short range order instead. However, since there is no clear distinction between anomalies due to the field induced order and those for the Schottky effect, in Fig. 1 we used one symbol for all data points for corresponding to \(x=0.1\). We would like to stress that the temperatures of the Schottky maximum in high and small fields are not changed between \(x=0\) and 0.1.

These Schottky anomalies are also seen in \(x=0.2\) for \(H=13\) and 14 T (Fig. 3). Similarly, the specific heat for \(x=0.6\) (not shown) displays broad structures 13 and 14 T, at the same roughly coinciding with those for \(x=0\) and 0.1, and 0.2, for the same fields. Thus, the La-doping does not change the CEF scheme and energies of Pr as postulated from the zero field investigation\([3]\). Therefore, the observed sensitivity of electronic properties of Pr\(_{1-x}\)La\(_x\)Os\(_4\)Sb\(_{12}\) on \(x\) have to be due to inter-site effects, most probably AFQ interactions. There is no AFQ transition for \(x=0.2\) (and 0.6; not shown) down to 0.4 K in H=10 T. Instead, the f-electron specific heat in-
FIGURE 1. Magnetic field phase diagram for x=0, 0.02, and 0.1. Closed and open circles for x=0 represent Schottky and AFQ anomalies, respectively.

creases continuously with a decrease of temperature. Recall that 10 T corresponds roughly to the crossing field for PrOs₄Sb₁₂ and the maximal AFQ transition temperature is somewhere between 9 and 10 T. The low temperature tail in Fig. 3 is due to nuclear effects.

Thus, the magnetic field study of Pr₁₋ₓLaₓOs₄Sb₁₂ provides additional evidences that single-ion parameters of Pr do not depend on the La-content. The field-induced AFQ is relatively robust but no signatures of this long range order can be detected in x = 0.2 and 0.6. An interesting question is the nature of the low temperature state in a crossing field for alloys with x > 0.2. What is the mechanism responsible for removal of entropy associated with a quasi-doublet? Very strong nuclear effects prohibited us to measure the specific heat at lower temperatures, with sufficient accuracy, for moderately diluted alloys.

ACKNOWLEDGMENTS

This work was supported by the Department of Energy under Grant No. DE-FG02-99ER45748.

REFERENCES