Anomalous Behavior in Quadrupolar Ordering Temperature in $Pr(Pb_{1-x}Sn_x)_3$

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Abstract

Specific heat measurements have been carried out in $Pr(Pb_{1-x}Sn_x)_3$ for $x = 0$, 0.03, 0.05, 0.1 and 0.2 with a non-Kramers Γ_3 doublet in the crystal-electric-field (CEF) ground state. The ordering temperature of the Γ_3 moment shows an anomalous behavior as Sn concentration increases. In $PrPb_3$ a sharp peak is observed at $T_Q = 0.4$ K, which is the quadrupolar ordering. For $x = 0.03$, the peak is suppressed below 0.1 K, while the peak shifts to higher temperatures with further increase of Sn concentration. For $x = 0.2$, the peak is observed at $\sim$0.2 K. The possible origin of this behavior will be discussed.

Key words: quadrupolar ordering; impurity effect; specific heat

1. Introduction

Some f-compounds which exhibit an antiferro-quadrupolar ordering (AFQPO) have attracted much attention because of the unique phase diagrams under magnetic fields and/or impurity substitutions. We have studied impurity effects on AFQPO in $PrPb_3$, and reported that AFQPO is expected to persist only up to $x \sim 0.02$ [1]. This impurity dependence is largely difference from those observed in $Ce_{1-x}La_xB_6$ [2] and $Dy_{1-x}Y_xB_2C_2$ [3]. We explained the difference by competition between the distortion of CEF due to the impurity ion and AFQPO temperature. The maximum distortion energy by La impurity is estimated to be of the order of 2$\sim$3 K in $PrPb_3$, which is much larger than AFQPO temperature $T_Q = 0.4$ K. If such a large distortion spreads around a La impurity over its third neighbors, the strong suppression can be explained.

When we see the impurity dependence of $Ce_{1-x}La_xB_6$ and $Dy_{1-x}Y_xB_2C_2$ from this point of view, these are well explained by the scenario. In $Ce_{1-x}La_xB_6$, the critical concentration of AFQPO is expected to be $x \sim 0.3$ from the phase diagram, while T_Q in CeB_6 is 3.3 K [2]. In $Dy_{1-x}Y_xB_2C_2$, the quadrupolar interaction is considered to be much larger than the distortion, because T_Q in DyB_2C_2 is 25 K. Therefore AFQPO survives up to $x = 0.6$ [3].

In the case of La substitution, however, the negative pressure effects exsit because the ion radius of La is larger than that of Pr. Thus we can not ruled out the possibility of the decrease of the quadrupolar interaction by increase of the volume. In order to examine our explanation for the substitution of the other ions and clarify the role of the volume change, we have studied the impurity effects of AFQPO in $PrPb_3$ by substituting Sn ions for Pb ions.

2. Experimental

All samples are prepared by the Bridgeman method. The susceptibility is measured down to 2 K in the static

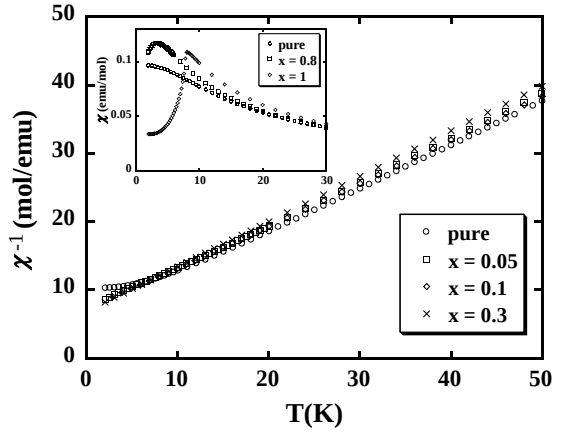

Fig. 1. Temperature dependence of the inverse susceptibility for pure and $x = 0.05$, 0.1 and 0.3 in $Pr(Pb_{1-x}Sn_x)_3$. Inset: Temperature dependence of the susceptibility for $PrPb_3$, $x = 0.8$ and $PrSn_3$. A sharp peak for $x = 0.8$ and $PrSn_3$ correspond to an antiferromagnetic ordering.

field of 1 kOe using a Quantum Design SQUID magnetometer. The specific heat is measured by a semiadiabatic method using a dilution refrigerator. A small amount of Apiezon grease is mixed into some pieces of sample crystals to keep good thermal contact.

3. Results

$PrPb_3$ and $PrSn_3$ have the $AuCu_3$-type cubic structure with $a = 4.903$ Å and 4.714 Å, respectively. Thus we could substitute Sn ions for Pb ions without the change of the crystal structure. The temperature dependence of the susceptibility for $PrPb_3$, $x = 0.8$ and $PrSn_3$ is plotted in the inset of Fig. 1. The susceptibility for $PrPb_3$ is well reproduced by CEF scheme with the ground state of Γ_3 and the first excited state of Γ_4 with the energy gap of 19 K. On the other hand, a peak due to the antiferromagnetic ordering (AFMO) is observed at $T_N = 8.6$ K for $PrSn_3$. The peak is depressed down to 4 K for $x = 0.8$.

The temperature dependence of the inverse susceptibility for the dilute region of Sn concentration with $x = 0.1$ and 0.2 and $PrPb_3$ is shown in Fig. 1. The susceptibility for $x = 0.1$ and 0.2 agree with that of $PrPb_3$ qualitatively in the temperature region above 4 K, which suggests that the CEF scheme is almost the same with that for $PrPb_3$. This is consistent with the previous experiment in $Pr(Pb_{1-x}Tl_x)_3$ by inelastic neutron scattering, in which the Γ_3 state in the CEF scheme maintains up to $x = 0.5$ despite the difference of the electronic charge that Pb and Tl ions are trivalent and tetravalent, respectively [4].

The temperature dependence of the specific heat for

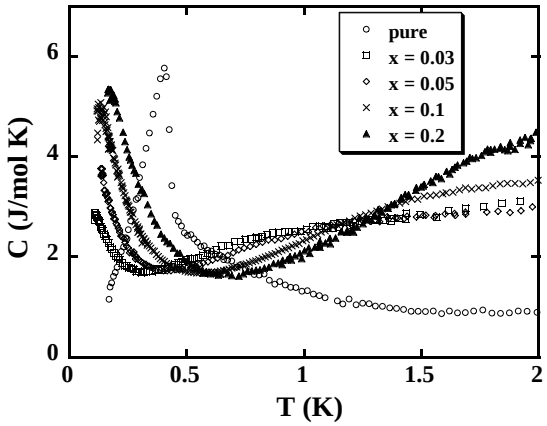

Fig. 2. Temperature dependence of the specific heat for $PrPb_3$ and the impurity system with $x = 0.03$, 0.05, 0.1 and 0.2.

$PrPb_3$ and Sn concentration with $x = 0.03$, 0.05, 0.1, and 0.2 is shown in Fig. 2. A sharp peak due to AFQPO is shown at $T_Q = 0.4$ K for $PrPb_3$. When Sn ions are substituted for Pb ions, the specific heat is largely modified from that for $PrPb_3$. The sharp peak is depressed below 0.15 K for $x = 0.03$, which can be explained as in the case of La substitution. With further increase of Sn concentrations, the peak shifts to higher temperatures. For $x = 0.2$, the peak is seen at $\sim$0.2 K. This concentration dependence of the peak is very curious, and can not be explained by the distortion of CEF. Two possible origins are offered for the shift of the peak. One is the enhancement of the quadrupolar-quadrupolar interaction due to the shrinkage of the lattice parameter, leading to increase of AFQPO. The other is the effect of AFMO attributed to Sn ions. Because the entropy change for the impurity system is larger than that for $PrPb_3$ below 2 K, suggesting the mixing of Γ_3 and the other state. Further experimental studies for the higher region of Sn concentration are needed to clarify the impurity effects of Sn.

4. Conclusion

We measured the specific heat in $Pr(Pb_{1-x}Sn_x)_3$ for $x = 0$, 0.03, 0.05, 0.1 and 0.2. A small amount of Sn ions gives the large influence for the low-temperature property in AFQPO.

References

[1] T. Kawae *et al.*, Phys. Rev. B **65** (2002) 012409.

[2] T. Tayama *et al.*, J. Phys. Soc. Jpn. **66** (1997) 2268.

[3] S. Sako *et al.*, J. Phys. Soc. Jpn. **69** (2000) 1872.

[4] G. Knopp *et al.*, J. Magn. Magn. Mater. **52** (1985) 326.