ELECTRONIC STRUCTURES OF $PrPb_3$ IN THE PARA- AND ANTIFERROQUADRUPOLAR PHASES*

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(Received July 10, 2002)

We have performed a detailed analysis of the dHvA oscillations in the para(PQ)- and antiferro-quadrupolar(AFQ) phases of $PrPb_3$. It is found that *(i)* each dHvA oscillation consists of up and down spin oscillations whose effective masses are very different, although the difference of the frequencies are small, *(ii)* the effective masses are largely enhanced compared to those of reference compound $LaPb_3$, and *(iii)* there is a strong anisotropy of the effective masses. The transition behavior of the dHvA oscillations across the phase boundary from AFQ to PQ phase is found to be also anisotropic. Particularly, a drastic change of the dHvA wave form and mass enhancement were observed for [001], suggesting a fluctuation of quadrupole and accompanying magnetic fluctuation.

PACS numbers: 79.60.–i, 71.18.+y

1. Introduction

$PrPb_3$ crystallizes into the $AuCu_3$-type cubic structure and exhibits antiferroquadrupolar transition at 0.4K. The CEF ground state is a non-Kramers doublet and no magnetic transition takes place. When a magnetic field is applied parallel to the [001] axis at low temperatures, the system enters paraquadrupolar(PQ) phase from antiferroquadrupolar(AFQ) phase at about 7T. The dHvA effect of $PrPb_3$ has been studied by Aoki *et al.* [1], and the effective masses and the angular dependence of the dHvA frequencies have been reported. We have found that each dHvA oscillation consists of two component oscillations whose frequencies are different from each other. From the analysis of the harmonic contents, we have proved that the two component oscillations originate from the up and down spin Fermi surfaces [2].

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

In this paper, we will report the effective masses of the up and down spin oscillations and their anisotropies as well as the anomalous behavior of the dHvA oscillations at the phase boundaries to discuss the effect of quadrupoles on the conduction electrons. To study the anisotropy, we have chosen the α and γ oscillations which arise from the spherical hole surfaces centered at the R and Γ points, respectively.

2. Experimental

We have grown the single crystals of $PrPb_3$ by the Pr self-flux method. The dHvA oscillations were detected from 1.2 K down to 50 K under magnetic fields up to 18 T using the conventional field modulation method.

Each dHvA oscillation was separately observed as a function of magnetic field at each temperature by using a filter.

3. Results and discussion

3.1. Effective masses

Table I shows the effective masses of the up and down spin oscillations and the relative difference of their frequencies for α and γ in the PQ phase. In the AFQ phase, it was difficult to determine the effective mass separately due to worse S/N ratio. Three remarkable features are found.

Firstly, the effective masses are much heavier than those of the reference compound $LaPb_3$. The maximum mass enhancement factor 11 is comparable to those of the dense Kondo compounds. It is not clear whether the mass enhancement can be thoroughly explained by the Kondo effect in $PrPb_3$ where the ground state is non-magnetic.

Secondly, the effective masses of the up and down spin oscillations are very different from each other and the maximum difference amounts to twice, although the difference in the two dHvA frequencies is small. This difference of the effective mass is much larger than those found in magnetic f electron compounds and is difficult to understand in terms of the spin splitting of the band.

Finally, the effective masses of α and γ depend strongly on the field direction. Since both the Fermi surfaces are approximately spherical, it is difficult to understand the anisotropy in terms of the anisotropy in the character of conduction band. We suppose that the anisotropy arises from the difference of the quadrupole state or f state which depends on the applied field direction and from the consequent difference in the interaction between conduction and f electrons.

TABLE I

Effective masses of up and down spin oscillations. Low Freq. and High Freq. denote the lower and higher frequencies of the two component oscillations, respectively. $\Delta F/F$ is the relative difference of the two frequencies.

γ oscillation

direction of H	[001]	[110]	[111]
Low Freq.	$2.6m_0$	$3.3m_0$	$2.4m_0$
High Freq.	$2.1m_0$	$5.3m_0$	$1.7m_0$
$\Delta F/F$	$\leq$0.02	0.027	$\leq$0.02
$PrPb_3$ [1]	$2.2m_0$	$2.0m_0$	$1.9m_0$
$LaPb_3$ [1]	$0.38m_0$	$0.46m_0$	

α oscillation

direction of H	[001]	[110]	[111]
Low Freq.	$2.3m_0$	$6.3m_0$	$4.2m_0$
			$3.9m_0$
High Freq.	$4.2m_0$	$4.1m_0$	$4.9m_0$
			$5.6m_0$
$\Delta F/F$	0.013	0.035	–
$PrPb_3$ [1]	$3.7m_0$	$6.7m_0$	$4.9m_0$
$LaPb_3$ [1]	$1.45m_0$	$1.46m_0$	

3.2. Phase transition and Fermi surface properties

Fig.1(a) and (b) show the γ oscillation for the fields parallel to the [110] and [001] directions, respectively. The solid lines indicate the phase boundaries. For [001], at least two phase boundaries are observed [3] and a metamagnetic behavior is also observed at the phase boundary. Both the oscillations form the beat structure indicating that the oscillation consists of two components.

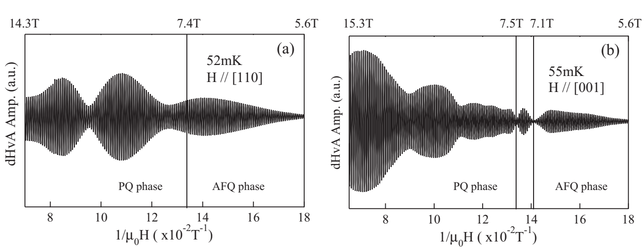

Fig. 1. dHvA oscillations of γ at low temperatures for fields parallel to [110] (a) and to [001] (b). The solid lines indicate the phase boundaries.

For [110], it is noted that the wave form changes smoothly in both the phases as well as across the phase boundary. The effective mass is also found to decrease gradually with increasing field in both the phases as well as at the phase boundary. The dHvA frequency is nearly the same in both the phases. A similar behavior is observed for [111].

For [001], the behaviors of wave form and effective mass are qualitatively similar to those for [110] and [111] in fields away from the phase boundaries. However, the wave form changes drastically near the phase boundary and the effective mass is found to be enhanced there [2]. The frequency is nearly constant as a function of magnetic fields except for the fields near the phase boundary where the frequency changes artificially due to the change of the wave form.

The phase transition is thought to be second order [3]. The smooth change across the phase boundary for [110] or [111] indicates a smooth change of the quadrupole or f electron states. On the other hand, we suspect that the anomalous behavior of the wave form and effective mass for [001] indicate the fluctuation of quadrupoles and accompanying magnetic fluctuations near the phase boundaries, although the reason why the fluctuation is strong only for [001] is not clear.

In summary we have observed very remarkable features of the Fermi surface properties in $PrPb_3$ which have not been observed in other materials. We think that these may be related with the quadrupole or the f electron state which depends on the field and its direction, and are also worth further intensive studies.

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