PHASE DIAGRAM AND dHvA EFFECT OF $PrPb_3$ UNDER PRESSURE*

M. Endo, N. Kimura, A. Ochiai, H. Aoki

Center for Low Temperature Science, Tohoku University, Sendai 980-8578, Japan

T. Terashima, C. Terakura, S. Uji
and T. Matsumoto

National Institute of Materials Science, Tsukuba, Ibaraki 305-0003, Japan

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We have studied the magnetic phase diagram and the de Hass-van Alphen (dHvA) effect of $PrPb_3$ under pressure. The antiferroquadrupolar (AFQ) transition temperature increases only slightly with pressure, whereas the transition field from AFQ phase to paraquadrupolar phase increases considerably with pressure. The effective masses of the α and γ oscillations increase largely with pressure implying that there is a significant change in the interaction or the hybridization between the f and conduction electrons. With the aid of the mean field analysis by Tayama *et al.*, we argue that the pressure strengthen the antiferromagnetic interaction, but does not affect the quadrupolar interaction appreciably.

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1. Introduction

$PrPb_3$ crystallizes into the $AuCu_3$-type cubic structure and exhibits antiferroquadrupolar transition at 0.4 K. 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] direction at low temperatures, the system enters paraquadrupolar (PQ) phase from antiferroquadrupolar (AFQ) phase at about 7 T.

The AFQ interaction is thought to be mediated via the conduction electrons. The pressure is expected to change the interaction between the quadrupoles and conduction electrons. Therefore, to investigate the inter-

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action, it will be useful to study the phase diagram under pressure together with the electronic structure change with pressure. We will report the de Hass–van Alphen (dHvA) effect and the magnetic phase diagram of $PrPb_3$ under pressure.

2. Experimental

We have grown single crystals of $PrPb_3$ by the Pb self-flux method. The high pressure is produced by a clamp type pressure cell [1]. The field was applied parallel to the [001] axis. The dHvA effect was measured under pressures up to 16.5 kbar and under magnetic fields up to 15 T using the conventional field modulation method.

3. Results and discussion

3.1. AC susceptibility and phase diagram

Fig. 1(a) and (b) show the AC susceptibility as a function of temperature and field at 16.5 kbar, respectively. The transition temperatures and fields are determined from the local maximum or inflection point as shown by the arrows. The transition at the highest field is broad and probably consists of two transitions [2]. We have observed a clear peak in the out of phase component of the AC susceptibility at the position of the arrow. In this paper we plot the position as the transition field of the highest field. Since there was a tiny amount of Pb flux on the surface of the sample, it was difficult to measure the AC susceptibility in zero field due to superconductivity of Pb.

Fig. 1(c) shows the magnetic phase diagram thus determined. The open and closed circles (triangles) denote the points determined from temperature and field sweeps at ambient pressure (16 kbar), respectively. The broken lines denote the phase boundaries at ambient pressure determined from the DC magnetization measurements by Tayama *et al.* [2]. The present result agrees well with their result. The solid lines are guides to the eye for the data points at 16.5 kbar. The same measurements were made at 3 kbar, 6 kbar, 9 kbar and 12 kbar, but are not shown for clarity. Another phase boundary is observed obviously at higher pressures, particularly under 16.5 kbar. The highest transition fields at lowest temperatures increase from about 7.5 T at ambient pressure to about 8.3 T at 16.5 kbar. The relative change is more than 10%. The AFQ transition temperatures in fields also increase with pressure. However, if we extrapolate the broken and solid lines to zero magnetic field, the AFQ transition temperature at 16.5 kbar does not seem to increase more than 20 mK from that at ambient pressure, *i.e.* the relative change is 5% at most.

Tayama *et al.*, [2] successfully explained the phase diagram by the mean field analysis. According to their analysis, the staggered moments are induced in fields and the antiferromagnetic interaction among them stabilizes the AFQ order to make the transition temperature increase with increasing field. Since there is no magnetic moment in the ground state, the present observation indicates that the pressure strengthens the antiferromagnetic interaction but does not affect the quadrupolar one appreciably.

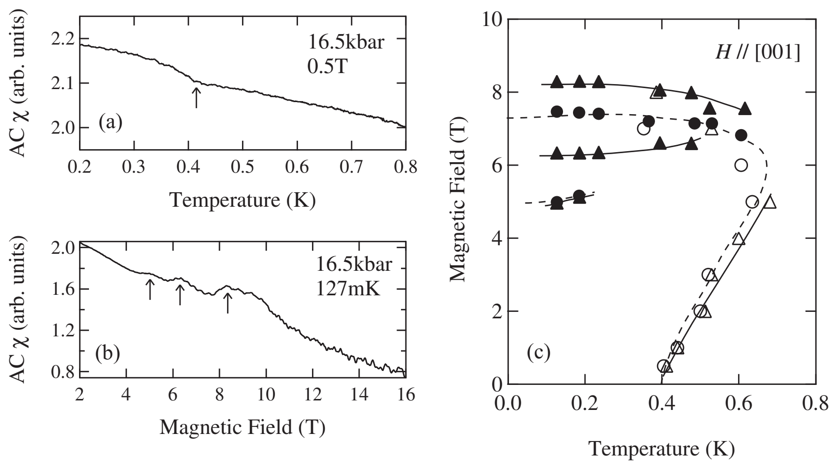

Fig. 1. (a) AC susceptibility under 16.5 kbar as a function of temperature at 0.5 T and (b) as a function of field at 127 mK. (c) Phase diagram of PrPb$_3$ for $H \parallel [001]$.

3.2. dHvA frequencies and effective masses

We have studied the pressure dependence of the dHvA frequency and effective mass for the α and γ oscillations which arise from the spherical hole surfaces at the R and Γ points, respectively. Fig. 2(a) shows pressure dependence of the dHvA frequency of γ. In PrPb$_3$, each dHvA oscillation consists of up and down spin oscillations whose frequencies are close to each other [3]. In this study, it is difficult to measure each oscillation separately due to worse S/N ratio of the measurements under pressure.

The frequency of γ decreases with increasing pressure up to 12 kbar and then seems to saturate at higher pressures. The decreasing rate of frequency up to 12 kbar is $2 \times 10^{-3}/$ kbar. This rate is about 10 times larger than that of normal metal Cu and is comparable to those of the localized f electron systems like CeB$_6$ and CeRu$_2$Ge$_2$ [4].

Fig. 2(b) shows the effective mass of γ as a function of pressure. The effective mass increases with increasing pressure up to 12 kbar and then starts to decrease. This behavior is consistent with the behavior of the frequency. The increasing rate up to 12 kbar amounts to 7×10^{-2}/kbar. This rate is comparable to that of a heavy fermion system $CeRu_2Si_2$ [4].

On the other hand, the frequency of α decreases with pressure, whereas the effective mass increases with pressure. The changing rates of the frequency and effective mass are comparable to those of γ.

The present results of the dHvA effect imply that the interaction between the f and conduction electrons changes considerably with pressure, probably through the change of hybridization.

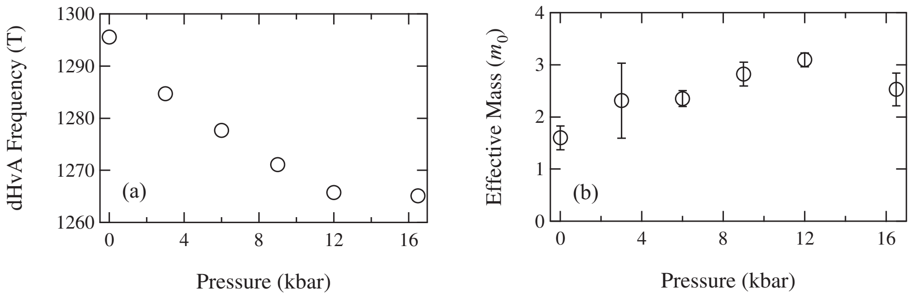

Fig. 2. (a) dHvA frequency of γ as a function of pressure. (b) Effective mass of γ as a function of pressure. The error bar indicates the standard derivation.

Both the antiferromagnetic and quadrupolar interactions are thought to be mediated via the conduction electrons. The present observations including that of the pressure effect on the phase diagram suggest that the quadrupolar interaction is less sensitive to the change in the hybridization between the f and conduction electrons than the antiferromagnetic interaction.

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