Quadrupolar Ordered State at High Fields in PrPb₃

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Abstract

PrPb₃ exhibits an antiferroquadrupolar transition at $T_Q=0.4$ K. In the present work, in order to reveal the order parameters of the ordered states in much higher fields than 5 T applied along the $[1\bar{1}0]$ direction, we performed neutron diffraction experiments in the temperature range between 50 mK and 0.8 K in magnetic fields up to 14.5 T. Above 7 T, we observed magnetic reflections at $\mathbf{q}=(\delta \delta 0.5)$ ($\delta \sim 0.02$) and its equivalents. The field-induced antiferromagnetic components were normal to the applied field, indicating that the quadrupolar order parameter in the high field phase should be of O_2^2 -type.

Key words: Antiferroquadrupolar ordering, Neutron diffraction

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1. Introduction

Antiferroquadrupolar (AFQ) ordering in the intermetallic f electron compounds has attracted much interest in these years. PrPb₃ crystallizes in a simple AuCu₃-type cubic structure, and has a Γ_3 non-Kramers doublet in the crystalline-electrical-field (CEF) ground state, with a magnetic Γ_4 triplet lying 15~19 K above the ground state.[1,2] Since the Γ_3 doublet carries quadrupole moments such as O_2^0 and O_2^2 , PrPb₃ is a good candidate for a quadrupolar transition. Actually, at the temperature of $T_Q=0.4$ K, a second order transition was observed with a lambda type anomaly in the spe-

cific heat measurement.[3,4] The idea of an AFQ ordering has further been strengthened by the $H-T$ diagram study[5], in which reentrant behavior with a significant enhancement of the transition temperature $T_Q(H)$ was observed for $H||\langle 100 \rangle$, as is often the case for AFQ ordering systems. Moreover, angle-resolved magnetization measurements revealed characteristic field-angular oscillations of $T_Q(H)$, which can be interpreted by assuming an O_2^0 -type AFQ moment and its equivalents to be the order parameters (OPs) at low $H(<6$ T).[6,7]

Very recently, we have revealed the quadrupolar structure in the AFQ phase by detecting an antiferromagnetic component induced by magnetic field applied along the $[001]$ direction.[8] Interestingly, the quadrupoles are distributed with the sinusoidally modulated structure, suggesting that the RKKY-type interaction between the quadrupoles plays an important role in this system.[9,10] Moreover, the direction of the field-induced magnetic

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moments is parallel to the magnetic field, indicating the quadrupolar OP in the AFQ phase should be of O_2^2 -type.[11] In much higher magnetic fields than 1 T, the sinusoidal structure turns into an antiphase structure with decreasing temperature below $T_t(<T_Q)$. On the other hand, when magnetic field was applied along the $[01\bar{1}]$ direction, no superlattice reflection was observed on the (hkk) scattering plane ($\perp \mathbf{H}$) below 3 T, although weak reflections at $\mathbf{q}=(0.5 \ \delta \ \delta)$ ($\delta \sim 0.062$) and its equivalents were observed only in the fields of 3 T $< H < 5$ T at 80 mK.[12] From the analysis of the $\mathbf{Q}$ -dependence of the reflection intensity, the field-induced component is determined to be perpendicular to the field, suggesting that the OP should be of O_2^2 -type.[11] The O_2^2 -type OP was also suggested from the measurements of the field angle dependence of the transition temperature; a new phase characterized by the transition temperature T_Q' was observed in fields above 7 T in narrow field directions around the $\langle 110 \rangle$ axis.[7] However, the ordered state in the magnetic fields above 5 T has not been revealed by a microscopic techniques yet.

In the present work, in order to reveal the OPs of the ordered states in much higher fields than 5 T applied along the $[1\bar{1}0]$ direction, we performed neutron diffraction experiment at temperatures ranging from 50 mK to 0.8 K and in magnetic fields up to 14.5 T. Above 7 T, we have found field-induced magnetic reflections at $\mathbf{q}=(\delta \ \delta \ 0.5)$ ($\delta \sim 0.02$) and its equivalents. The $\mathbf{Q}$ -dependence of their intensity indicates that the field induced antiferromagnetic component is perpendicular to the field, confirming that the quadrupolar OP of this new ordered phase should be of O_2^2 -type. In the fields of 3 T $\leq H \leq 7$ T, we have also observed some reflections, for example, at $\mathbf{q}=(\delta \ \delta \ 0.5)$ ($\delta \sim 0.062$) and its equivalents which were reported before.[12]

2. Results and Discussion

A single crystalline sample of PrPb₃ was grown by the Bridgman method using a Mo crucible. In the present study, a specimen with dimensions of $10 \times 8 \times 5$ mm³ mounted on a mixing chamber of a ³He–⁴He dilution refrigerator (base temperature 50 mK) was placed in a vertical superconducting cryomagnet capable to generate 14.5 T. The magnetic field was applied along the $[1\bar{1}0]$ direction, therefore, the scattering plane was the (hhl) plane perpendicular to the magnetic field. Neutron diffraction

measurements were performed using the double-axis spectrometer E4 installed at the HMI with incident neutron wave length of 2.44 Å.

Figure 1 shows the $\mathbf{Q}$ -scan along the $(h \ h \ 0.5)$ line at the temperature of $T=0.1$ K and in various magnetic fields of $0 \leq H \leq 14.5$ T. Below 3 T, we could not observe any magnetic reflections neither on the $(h \ h \ 0.5)$ line nor along the principal lines on the (hhl) plane such as the $(hh0)$, $(00l)$ and (hhl) lines. Increasing the magnetic field above 7 T, magnetic reflections appeared at $\mathbf{q}=(\pm\delta \ \pm\delta \ 0.5)$ ($\delta \sim 0.02$) and their equivalents. The intensity increases with increasing H from 7 T to 10 T, but turns to decrease at higher fields. Even at the maximum field of 14.5 T, however, the reflections still remain at 0.1 K. We also observed their third harmonic reflections at around 10 T. They probably result from squaring-up of a modulated structure as will be shown later. We observed the small superlattice reflections at $\mathbf{q}=(\delta \ \delta \ 0.5)$ ($\delta \sim 0.062$) and their equivalents in the fields of 3 T $\leq H \leq 7$ T. This result confirmed our previous data obtained at $T=40$ mK and in the magnetic fields of 3 T $\leq H \leq 5$ T.[12] Moreover, in the field of 6 T $< H < 7$ T, we observed other superlattice reflections in the $(h \ h \ 0.5)$ line. Although we are not able to determine the structures from the reflections because they are very small and undefined, they probably result from instability of the OPs shown as follow.

In order to confirm the phase boundary between the AFQ ordered and the paramagnetic states, we examined the temperature dependence of the intensity of the magnetic reflections. Figure 2 shows the evolution of the integrated intensity of the magnetic reflection at $\mathbf{q}=(-\delta \ -\delta \ 0.5)$ ($\delta \sim 0.02$) in the magnetic field of $H=10$ T. The dotted line shows the background of the intensity. The intensity decreases continuously with increasing T , and it diminished to the back ground level at the transition temperature of $T_Q'=0.58$ K as shown with the thick arrow in Fig. 2, indicating that the ordered state collapses above T_Q' . We also examined the temperature dependence of the intensity in various fields. The transition temperature T_Q' varies with changing the magnetic fields. From the results, we confirmed that the field dependence of T_Q' thus obtained was in good agreement with the H – T phase diagram determined by our magnetization measurements.[7] On the other hand, the boundary of the ordered phase characterized by the appearance of the magnetic reflection at $\mathbf{q}=(\delta \ \delta \ 0.5)$ ($\delta \sim 0.062$) could not be clearly defined because of the weakness of the intensity.

The magnetic structure in the ordered phases can be derived from the $\mathbf{Q}$ -dependence of the intensity of the magnetic reflections. Figure 3 shows the antiferromagnetic components of the field-induced magnetic structure determined from the magnetic reflections at $\mathbf{q}=(\pm\delta \pm\delta 0.5)$ ($\delta\sim 0.02$) and their equivalents at $T=0.1$ K and in the magnetic field of $H=10$ T. We found that the intensity of the reflection at $\mathbf{q}=(-\delta -\delta 0.5)$ was twice as large as the one at $\mathbf{q}=(\delta \delta 0.5)$. In the ideal condition, the intensity of these two peaks should coincide with each other. The disparity could possibly be attributed to a misalignment of the field direction. We therefore took the average of these satellite intensity for analysis of the magnetic structure. We assumed that antiferromagnetic component is parallel to the $[110]$ direction and is modulated with the propagation vector of $\mathbf{k}=[\pm\delta \pm\delta 0.5]$ ($\delta\sim 0.02$). Fig. 3 shows the alignment of the moment in the $z=0$ plane. The direction of each moment changes alternatively along the $[001]$ direction. We determined the amplitude of the sinusoidal structure to be $0.93(15) \mu_B$. The fact that the antiferromagnetic component is perpendicular to the field suggests that the quadrupolar OP in the ordered phase should be the O_2^2 -type.[11] The uniform component of the magnetic moments at 10 T estimated from the field-enhanced intensity of the nuclear Bragg peaks was $1.52(20) \mu_B$, being consistent with the results of the magnetization measurements.[7]

We now discuss the possible OPs in the ordered phases. In the magnetic field applied along the $\langle 100 \rangle$ direction, the OP has been confirmed to be of O_2^0 -type by our previous neutron diffraction experiment in magnetic fields, where the antiferromagnetic component of the field-induced magnetic moments is parallel to the field.[8] This is consistent with the OP suggested by the angular dependence of the transition temperature T_Q . [6] The quadrupolar ordering temperature T_Q varies with rotating the field. T_Q has a maximum value in the $\langle 100 \rangle$ direction and a cusp-type minimum in the $\langle 110 \rangle$ direction below 4 T. These results were well explained by assuming that the low field phase consists of O_2^0 -type OP with three domains as explained in detail in Ref. [6]. On the other hand, in the angle-resolved magnetization measurements performed at higher fields, T_Q appeared in the fields above 7 T in a narrow angular range very close to the $\langle 110 \rangle$ axis.[7] Considering that the Γ_3 doublet carries the quadrupoles O_2^0 and O_2^2 , the OP of the ordered phase observed around the $\langle 110 \rangle$ direction above 7 T was expected to be

of O_2^2 -type. This point has strongly been supported by the present neutron experiment in which the antiferromagnetic component of the $\langle 110 \rangle$ high field phase is perpendicular to the field, being compatible with the O_2^2 AFQ OP.[11] The present neutron data, combined with our magnetization and neutron experiments, indicates that the OP changes from O_2^0 to O_2^2 types in magnetic fields along the $\langle 110 \rangle$ direction. This change of the OPs in magnetic fields would probably result from the anisotropic interaction between the quadrupoles, where the O_2^0 type OP is preferred over the O_2^2 one in zero field. This was also pointed out by the angle-resolved magnetization measurements.[6] The complicated and strongly anisotropic phase diagrams of PrPb_3 strongly suggests the relevance of the anisotropic quadrupolar interaction in this system.

3. Conclusion

In summary, we performed the neutron diffraction experiments on PrPb_3 in the magnetic fields up to 14.5 T applied along the $\langle 110 \rangle$ direction. Above 7 T, the magnetic reflections were observed at $\mathbf{q}=(\pm\delta \pm\delta 0.5)$ ($\delta\sim 0.02$) and its equivalents. In the field of $3 \text{ T} < H < 7 \text{ T}$, the weak reflections were observed, for example, at $\mathbf{q}=(\pm\delta \pm\delta 0.5)$ ($\delta\sim 0.062$) and its equivalents. On the other hand, no superlattice reflection was observed below 3 T in the (hhl) scattering plane. The quadrupolar OP in the ordered phase above 7 T, characterized by the transition temperature T_Q , was confirmed to be of O_2^2 -type, which is consistent with the results of the angle-resolved magnetization measurements. The results suggest that the OP should change from the O_2^0 -type to the O_2^2 -type by applying the magnetic field along the $\langle 110 \rangle$ direction. This behavior of the OPs and the anisotropic and complex phase diagrams probably result from the anisotropic RKKY-type interaction between the quadrupoles.

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Fig. 1. Q -scan along the $(h h 0.5)$ -line at the temperature of $T=0.1$ K in the magnetic fields up to 14.5 T applied along the $[1\bar{1}0]$ direction. Each curve is vertically shifted for clarity.

Fig. 2. Temperature dependence of the integrated intensity of the field-induced magnetic reflection at $q=(\delta \delta 0.5)$ ($\delta \sim 0.02$) in the magnetic field of $H=10$ T applied along the $[1\bar{1}0]$ direction. The dotted line shows the background of the intensity. The thick arrow indicates the transition temperature T'_Q .

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Fig. 3. Field-induced magnetic structure determined by the Q dependence of the intensity of the magnetic reflection at $q=(\pm\delta \pm\delta 0.5)$ ($\delta \sim 0.02$) and their equivalents at the temperature of $T=0.1$ K and in the magnetic field of $H=10$ T. The antiferromagnetic component is parallel to the $[110]$ direction and the components are modulated with the amplitude of $\mu_{AF}=0.93(15) \mu_B$ and the propagation vector of $k=[\pm\delta \pm\delta 0.5]$ ($\delta \sim 0.02$). The magnetic moments are distributed alternately along the $[001]$ direction.

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