(7c9)

Modulated Quadrupole Ordering Structures in $PrPb_3$

T. Onimaru[1], T. Sakakibara[1], N. Aso[2], H. Yoshizawa[2], H. Suzuki[3] and T. Takeuchi[4]

[1]Institute for Solid State Physics, University of Tokyo, Kashiwa, 277-8581
[2]Neutron Science Laboratory, Institute for Solid State Physics, University of Tokyo, Tokai 319-1106
[3]National Institute of Materials Science, Tsukuba 305-0047
[4]Low Temperature Center, Osaka University, Toyonaka 560-0043

In the present study, we concentrate our attention on the intermetallic compound$PrPb_3$ with $AuCu_3$-type cubic structure. The crystalline field ground state of $PrPb_3$ is a Γ_3 non-Kramers doublet, with a magnetic Γ_4 triplet lying 15∼19 K above the ground state.[1-3] Since the Γ_3 doublet carries quadrupolar moments $O_2^0=(3J_z^2-\boldsymbol{J}^2)/2$ and $O_2^2=\sqrt{3}(J_x^2-J_y^2)/2$, $PrPb_3$ is a good candidate for the quadrupole transition. This compound actually exhibits a second-order transition at 0.4 K with a lambda-type anomaly in the specific heat.[4,5] Absence of a magnetic superlattice reflection nor a lattice distortion in the neutron diffraction measurement performed in zero magnetic field[1] suggests the phase transition to be of AFQ.

We performed neutron diffraction measurements on the cubic compound $PrPb_3$ in a [001] magnetic field to examine the quarupolar ordering. Fig. 1 shows the results of $\boldsymbol{Q}-$scans along the $(h\frac{1}{2}0)$ line carried out in a field of H=4 T at various temperatures ranging from 0.125 K to 0.8 K. The inset of Fig. 1 shows the $(hk0)$ reciprocal plane ($\perp H$) investigated, where open and closed circles represent the nuclear and the magnetic reflections, respectively, observed in a field of H=4 T at T=0.125 K. On cooling below the transition temperature T_Q=0.65 K, superlattice reflections with $\boldsymbol{q}=(\frac{1}{2}\pm\delta\frac{1}{2}\ 0)$, $(\frac{1}{2}\frac{1}{2}\pm\delta\ 0)$, $(\delta\sim\frac{1}{8})$ are observed. These reflections show that quadrupoles are ordering with a sinusoidal modulated structure. The intensity of these reflections vary linear to H and vanish at zero field, providing the first evidence for a modulated quadrupolar phase. For $H<1$ T, a non-square modulated state persists to below 100 mK suggesting quadrupole moments associated with a Γ_3 doublet ground state to be partially quenched by hybridizations with conduction electrons. On further cooling below T_t=0.45 K, the third-order harmonics $(\frac{1}{2}\pm3\delta\frac{1}{2}\ 0)$ and $(\frac{1}{2}\frac{1}{2}\pm3\delta\ 0)$ with much weaker intensity is found to develop. This state undergoes a first-order transition to an antiphase structure.

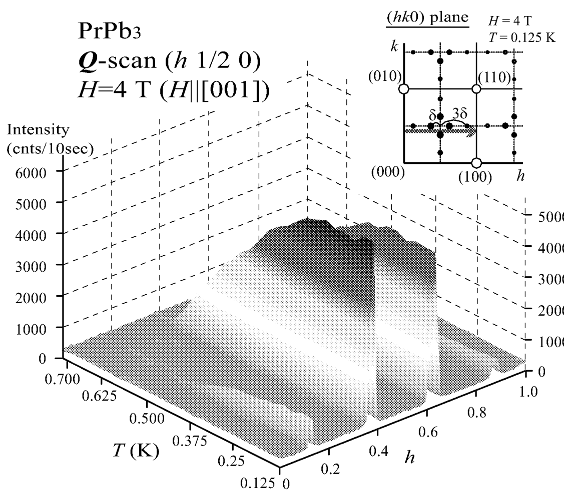

Figure 1: Evolution of the magnetic scattering in a field of H=4 T applied along the [001] direction, obtained in a temperature interval of 0.125 K<T<0.81 K. The $\boldsymbol{Q}-$scans were performed along the line $(h\frac{1}{2}0)$, as indicated by a arrow in the $(hk0)$ reciprocal plane (inset) where open and closed circles represent the nuclear and the magnetic reflections, respectively, observed at T=0.125 K.

Ref.
[1] M. Niksch et al., Helv. Phys. Acta 55, 688 (1982).
[2] W. Gross et al., Z. Phys. B 37, 123 (1980).
[3] T. Tayama et al., J. Phys. Soc. Jpn. 70, 248 (2001).
[4] E. Bucher et al., J. Low Temp. 2, 322 (1972).
[5] D. Aoki et al., J. Phys. Soc. Jpn. 66, 3988 (1997).