Phase Transitions of Non-Kramers Pr Ions in a Trigonal Symmetry in $Pr_4Ni_3Pb_4$

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Multipolar degrees of freedom often play an important role in $4f$ electron systems. Non-Kramers ions such as Pr^{3+} and Tm^{3+} possess multipolar degrees of freedom other than magnetic dipoles even in a trigonal symmetry. In the present work, we have focussed on a Pr-based intermetallic compound $Pr_4Ni_3Pb_4$ crystallizing in the trigonal $La_4Ni_3Pb_4$-type structure (space group $R3$) without inversion symmetry.[1, 2] In the unit cell, Pr ions occupy the $3a$ and $9b$ sites with the C_3 and C_1 point symmetries. In the C_3 point symmetry, a nine-fold multiplet $^3H_4$ splits into three Γ_1 singlets and three Γ_{23} doublets with quadrupolar degrees of freedom. The isothermal magnetization shows a shoulder-like anomaly at 4 T only for $B||c$, suggesting that an excited Γ_{23} doublet exists at a low energy of 4 K above the Γ_1 singlet ground state. The specific heat has cusp-type double anomalies at T_{N1}=2.7 K and T_{N2}=2.1 K which are probably attributed to the Γ_1-Γ_{23} quasi-triplet.

Neutron diffraction measurements on a single crystalline sample were performed using the ISSP High Q-resolution Triple-Axis Spectrometer HQR(T1-1), in order to detect magnetic peaks below the transition temperatures. Figure 1 shows the Q-scan along the $(1,0,L)$ line. A magnetic peak appears at $Q=(1,0,\frac{1}{4})$ and its equivalent positions below T_{N2}=2.1 K. At the temperature range between T_{N2} and T_{N1}, the peak at $(1,0,\frac{1}{4})$ splits into two peaks at $(1,0,\frac{1}{4}\pm\delta)$ ($\delta\sim0.1$) where positions shift on cooling. These behaviors indicate an incommensurate magnetic structure. We observed both peaks at 2.14 K close to T_{N2}, suggesting that the IC-C transition at T_{N2} should be of first order. Comparing the intensities of the equivalent magnetic peaks, they tend to be strongly suppressed as the peak positions approach to the [001]* direction. It means that the magnetic moments have a trend to align along the c-axis in the ordered structures. Detailed analyses of the magnetic structures are in progress.

References

[1] L. D. Gulay *et al.*: J. Alloys Compd. **392** (2005) 165.

[2] K. Shigetoh *et al.*: J. Phys. Soc. Jpn. **75** (2006) 033701.

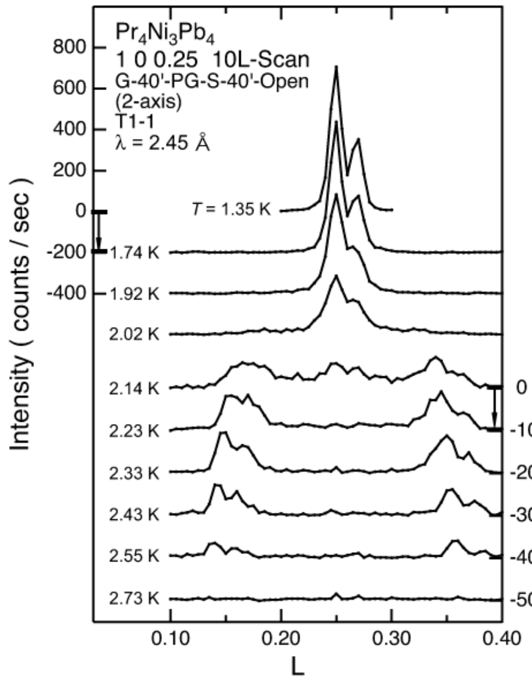

Fig. 1. Q-scan along the $(1,0,L)$ line. The magnetic $10\frac{1}{4}$ peak appears below T_{N2}=2.1 K.