# Multipolar Transition in a Trigonal $Pr_4Ni_3Pb_4$ with Non-Kramers Ground State

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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 $Pr_4Ni_3Pb_4$, where Pr ions occupy the $3a$ site with the $C_3$ point symmetry and the $9b$ site with the $C_1$ point symmetry. In the $C_3$ point symmetry, a nine-fold multiplet $^3H_4$ splits into three $\Gamma_1$ singlets and three $\Gamma_{23}$ doublets with quadrupolar degrees of freedom, although, in the $C_1$ point symmetry, nine $\Gamma_1$ singlets. The isothermal magnetization shows a shoulder-like anomaly at 4 T only for $\boldsymbol{B}||\boldsymbol{c}$, suggesting that an excited $\Gamma_{23}$ doublet exists at a small energy of 4 K above the $\Gamma_1$ singlet ground state of the Pr at the 3a site. The specific heat has cusp-type double anomalies at $T_{N1}$=2.7 K and $T_{N2}$=2.1 K.

In neutron diffraction measurements on a single crystalline sample, 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\sim$0.1) where the peak 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.

Figure 1 shows the inelastic neutron scattering spectra of $Pr_4Ni_3Pb_4$ at $T$=4 K and 30 K, and that of the reference $La_4Ni_3Pb_4$ at $T$=4 K. At 2.5 meV and 6.7 meV, peaks were observed. The intensities of the peaks decrease with increasing temperature, therefore, they result from crystalline electric field (CEF) excitations. These are also confirmed because no peak was observed at the energies in the reference $La_4Ni_3Pb_4$. From the CEF analysis on the magnetization, the peak at 2.5 meV is probably ascribed to a CEF excitation at the Pr at the 9b site with the $C_1$ point group, however, the peak at 6.7 meV the Pr at the 3a site with the $C_3$ point group. We now analyses the temperature dependence of the intensities of the CEF excitation peaks to confirm the CEF level scheme of both the Pr sites in $Pr_4Ni_3Pb_4$.

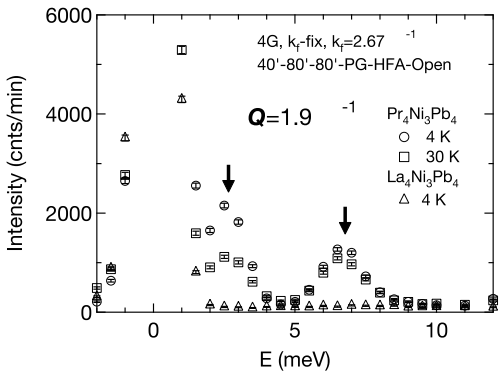


Fig. 1. Inelastic neutron scattering spectra of $Pr_4Ni_3Pb_4$ at $T$=4 K and 30 K, and that of the reference $La_4Ni_3Pb_4$ at $T$=4 K.