(28b6)

Multipolar Moments in Pr-based Filled Skutterudites with Singlet-Triplet Crystal-Field Levels

R. Shiina

Graduate School of Science, Tokyo Metropolitan University, Hachioji, 192-0397

Pr ions in filled skutterudites are believed to have a $4f^2$ configuration with a total angular momentum $J = 4$. The site symmetry is T_h, which is characterized by the absence of the four-fold symmetry axes. After some controversies, the crystal-field (CF) level scheme of $PrOs_4Sb_{12}$ is established as follows: The Γ_1 singlet is the CF ground state and a triplet, usually denoted as $\Gamma_4^{(2)}$, is located at $\sim$ 8K above the ground state. The singlet-triplet scheme is also a candidate of the CF model for other materials such as $PrFe_4P_{12}$ and $PrRu_4Sb_{12}$. Thus, one can expect that this level scheme is a common feature of Pr ions in the skutterudite compounds.

In this work, we analyze the multipoles in the singlet-triplet subspace, and discuss mainly their general aspects, using the analogies with two well-known systems. First, considering a similarity with pure quartet systems, one can relate the present problem with multipolar physics in CeB_6. This relationship is useful especially for the classification of multipoles and the analysis on their symmetry properties. It is shown that there are a larger number of nonmagnetic multipoles than those in CeB_6, which contains higher-rank ones, the hexadecapoles. Second, it is worth noting that the level scheme is the same as that of spin dimer systems. A pseudo-spin description considering this analogy makes clear the algebraic nature of multipoles and some hidden symmetry. Combining these analyses, we construct a generic form of multipolar interactions, which is used to identify the sources of cubic anisotropy for directions of magentic field.

Based on these considerations, the properties of quadrupolar interaction in the singlet-triplet system are studied in connection with the field-induced ordered state in $PrOs_4Sb_{12}$. The phase diagram for temperature vs. magnetic field is presented for several field directions, making use of the mean-field approximation. It is pointed out that the T_h symmetry specific to the filled-skutterudite systems is very important for the anisotropic phase diagram of $PrOs_4Sb_{12}$. A possible effect of magnetic multipoles is briefly discussed.

[1] R. Shiina and Y. Aoki: J. Phys. Soc. Jpn. **73** (2004) 541.
[2] R. Shiina: J. Phys. Soc. Jpn. **73** (2004) No. 8.

Table I. Active multipoles in O_h.

symmetry	$d = 0$ (Γ_1-Γ_5)	$d = 1$ (Γ_1-Γ_4)
Γ_4^-	$\boldsymbol{J}$	$\boldsymbol{J}$, $\boldsymbol{T}^{\alpha}$
Γ_5^-	$\boldsymbol{T}^{\beta}$	–
Γ_1^+	H^0	H^0
Γ_3^+	(O_2^0, O_2^2)	(O_2^0, O_2^2)
Γ_4^+	–	$\boldsymbol{H}^{\alpha}$
Γ_5^+	$\boldsymbol{O}^{\beta}$, $\boldsymbol{H}^{\beta}$	$\boldsymbol{O}^{\beta}$

Table II. Active multipoles in T_h.

symmetry	$d \approx 0$ (Γ_1-$\Gamma_4^{(2)}$)	$d \approx 1$ (Γ_1-$\Gamma_4^{(1)}$)
Γ_4^-	$\boldsymbol{J}$, $\boldsymbol{T}^{\beta}$	$\boldsymbol{J}$, $\boldsymbol{T}^{\alpha}$
Γ_1^+	H^0	H^0
Γ_3^+	(O_2^0, O_2^2)	(O_2^0, O_2^2)
Γ_4^+	$\boldsymbol{O}^{\beta}$, $\boldsymbol{H}^{\beta}$	$\boldsymbol{O}^{\beta}$, $\boldsymbol{H}^{\alpha}$

$\boldsymbol{J}$, $\boldsymbol{O}$, $\boldsymbol{T}$ and $\boldsymbol{H}$ denote three-component vectors; $\boldsymbol{O}$ is quadrupole, whereas $\boldsymbol{T}$ and $\boldsymbol{H}$ denote octupole and hexadecapole, respectively.