Magnetic excitations of Er$_2$PdSi$_3$ studied by inelastic neutron scattering in fields up to 12 T

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Abstract. The series $R_2$PdSi$_3$ ($R =$ heavy rare earth) has been found to exhibit rich magnetic phase diagrams with a large variety of magnetic states. This complex magnetic behavior results from the interplay between RKKY interaction, magneto-crystalline anisotropy based on crystalline-electric field (CEF) effects and geometric frustration due to the AlB$_2$ derived hexagonal crystal structure. Within the series the Er$_2$PdSi$_3$ compound has been found to exhibit in zero field a simple CEF excitation spectrum with just one strong transition at 3.5 meV from the ground state to the first excited state. Neutron spectroscopy in magnetic fields up to 12 T has been performed on the cold-triple axis spectrometer PANDA at FRM II. In this study two transitions within the 3.5 meV excitation could be resolved showing a linear dependency in applied magnetic fields with an almost identical slope of $\mu_B g_L$ for fields above 2 T. The results can be explained in a picture of two slightly different sets of CEF parameters for two inequivalent Er$^{3+}$ sites due the crystallographic superstructure observed in all single crystals of type $R_2$PdSi$_3$.

1. Introduction

The series of $R_2$PdSi$_3$ ($R =$ rare earth) compounds has been a focus of interest for over 15 years$^1$. The compounds crystallize in an AlB$_2$-derived hexagonal structure (P6/mmm) with a latent geometric frustration. The magnetic rare-earth ions occupy the Al positions of the AlB$_2$ structure while the non-magnetic Pd and Si atoms are assumed to be statistically distributed on the B positions$^2$. However, recent studies on single crystalline samples in the series revealed that Pd and Si atoms are forming some kind of ordering on the B sites instead of being randomly distributed$^3$. This crystallographic superstructure can be characterized by the extra reflections at Q positions (1/2, 1/2, n/8) with n being integer. Consequently, the unit cell is doubled in the basal plane and octupled along the c axis resulting in multiple inequivalent CEF environments for the rare earth ions.

It was previously assumed that the magnetic properties of the compounds are not influenced by the crystallographic superstructure. However, preliminary studies indicate that the crystallographic superstructure is closely related to a generic magnetic structure observed in applied magnetic field for most of the compounds in the series. Thus it is very intriguing to ask the question: how does the crystallographic superstructure influence the magnetic properties of the system?
2. Experimental details
The single crystal preparation and characterization are described elsewhere[4, 5]. The inelastic neutron scattering experiments were carried out on the cold triple axis spectrometer PANDA at the FRM II in Garching. The Er$_2$PdSi$_3$ (m ≈ 1300 mg) single crystal used in this study was of high quality. It was mounted in a 15 T vertical magnet with the [001] axis parallel to the field direction making the $ab$ plane the scattering plane. All measurements have been performed at 1.5 K. The spectrometer was operated in its fully focused mode at constant $k_f = 1.55 \AA^{-1}$. A liquid Nitrogen cooled Be filter was mounted before the analyzer to suppress the $\lambda/2$ contamination.

3. Crystal Electrical Field Effect in Er$_2$PdSi$_3$

The Er$^{3+}$ ion has a total angular momentum of $15/2$ with the Landé factor $g_L = 1.2$. The compound orders at $T_N = 7.0$ K with the magnetic moment parallel to the [001] direction and the propagation vector $\tau = (1/9, 1/9, 0)$ in the basal plane[6]. At a relative low critical field (1.2 T along [001] direction at 2 K) the compound is already in the induced ferromagnetic state with the fully saturated magnetic moment on the Er$^{3+}$ ion[7]. In this state the RKKY exchange interaction is negligible compared to the external field and the CEF.

For a hexagonal crystal structure, there are four non-zero $B_{m}^{n}$ CEF parameters in the CEF Hamiltonian: $H_{CEF} = B_{2}^{0}O_{2}^{0} + B_{4}^{0}O_{4}^{0} + B_{6}^{0}O_{6}^{0} + B_{8}^{0}O_{8}^{0}$ with the Stevens operators $O_{m}^{n}[8]$. Consequently, for Er$_2$PdSi$_3$ there are 8 doublets with wave functions of the form $|\psi\rangle = a|M_{J} + 6\rangle + b|M_{J} - 6\rangle$ where $|M_{J}\rangle$ is the eigenfunction of the angular momentum operator $J_z$. The magnetization curve of Er$_2$PdSi$_3$ suggests that the Er$^{3+}$ ion reaches its saturation moment (9 $\mu_B$/ion) at 1.2 T when an external magnetic field is applied along the [001] direction[6]. This implies that the CEF ground state wave function must be of the form: $|\psi_0\rangle = a|15/2\rangle + b|3/2\rangle + c|9/2\rangle$. In magnetic fields $\mu_0H > 1.2$ T there should be only one CEF transition for neutron scattering from the ground state $|\psi_0\rangle$ to the excited state $|\psi_1\rangle = a|13/2\rangle + b|1/2\rangle + c|11/2\rangle$ with an additional energy transfer proportional to the Zeeman energy of $\mu_B \cdot g_L \cdot \mu_0H$.

4. Results

4.1. Field dependence of CEF excitations
In the measurements for magnetic fields above 1.2 T we could resolve two peaks for the CEF transition around the energy transfer at 3.5 meV. Figure 1 shows the energy spectra obtained at $Q = (0.0, -1.0, 0.0)$ in the reciprocal space at $T = 1.5$ K with $\mu_0H = 2$ T and 12 T. The two transitions have an energy difference of around 0.3 meV. In figure 2 we present the field dependence of the positions of the two transitions for fields between 0 T to 12 T. In the field range above 2 T the energy transfer of the two transitions increases linearly with field with almost the identical rate of $\sim 0.07$ meV/T which has to be compared to the theoretical value $\mu_B \cdot g_L = 0.0695$ meV/T. Thus both transitions are connecting the ground state $|\psi_0\rangle$ with the first excited state $|\psi_1\rangle$ as given in the previous section.

This suggests that the reason for the two transitions must be coming from two different Er$^{3+}$ sites with slightly different local environments. This agrees with the findings about the crystallographic superstructures in the series: the superstructure with an enlarged unit cell of the size (2x2x8) times the original unit cell could produce more than one local environment for the rare earth site. Furthermore, preliminary results for the superstructure suggest one hexagonal and three monoclinic symmetric sites. This ratio roughly agrees with the ratio between the intensities of the two transitions suggesting that the 3.6 meV transition at 2 T in figure 1 originates from the hexagonal sites and the 3.3 meV transition from the monoclinic sites.
Figure 1. Energy spectra for Er$_2$PdSi$_3$ obtained at Q = (0.0, -1.0, 0.0) at T = 1.5 K: blue • and black ■ symbols are the experimental data points for 2 T and 12 T, respectively. Full lines are total fits for both transitions, dashed lines indicate the individual contributions.

4.2. Directional dependence of CEF excitations
To further elucidate the origin of the two CEF transitions, measurements have been made on its Q dependence in the reciprocal a$^*$b$^*$ plane. The intensity of the two transitions shows a strong directional dependence between the (H, 0, 0) and (H, H, 0) directions. Shown in figure 3 are the energy spectra taken at the Q positions (0, -1, 0) and (-0.5, -0.5, 0) at 1.5 K and 2 T with the field direction parallel to the [001] direction. The intensity of the second transition at higher energy transfer is almost directional independent while the intensity of the first transition at lower energy transfer almost doubled going from (-0.5, -0.5, 0) to (0, -1, 0).

The neutron scattering cross section of a CEF transition is dependent on the transition matrix element of angular momentum operator $J_\alpha$ where $\alpha = x, y, z^{[9]}$. For hexagonal symmetry with CEF wave functions of the form $|\psi> = \sum_{|M_J+6i|\leq J} a_i|M_J+6i> \cdot \mu_B \cdot g_i$. Taking into account the crystallographic superstructure which has been observed in the series $R_2$PdSi$_3$, the two CEF transitions can be

5. Conclusions
Detailed inelastic neutron scattering measurements have been performed on Er$_2$PdSi$_3$ to examine the magnetic excitations in the compound. In the induced ferromagnetic phase two CEF transitions have been observed while only one is expected in a simple AlB$_2$ hexagonal structure. The energy transfers of the two transitions increase linearly with the rate 0.07 meV/T for applied magnetic fields along the [001] direction when the field is larger than 2 T. This is in excellent agreement with a calculated slope of $\mu_B \cdot g_i$. Taking into account the crystallographic superstructure which has been observed in the series $R_2$PdSi$_3$, the two CEF transitions can be
attributed to two inequivalent rare earth sites surrounded by different distributions of Pd and Si atoms on the B positions.

Moreover, the intensity of the first transition at lower energy transfers shows a strong directional dependence with a factor around two between (0.0, -1.0, 0.0) and (-0.5, -0.5, 0.0) positions while the second transition is almost directional independent. This highly suggests that the local environment for the first transition is monoclinic while the local environment for the second transition has still hexagonal symmetry.

The results demonstrate that although the crystallographic superstructure is a relative weak modification of the major crystal structure, its influence through the modification of the CEF on the magnetic properties of the $\text{R}_2\text{PdSi}_3$ compounds is not negligible. More experiments are planned to examine the magnetic excitations of other members of the $\text{R}_2\text{PdSi}_3$ series to further understand the effects of the crystallographic superstructure on the magnetic properties of these compounds.

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