A Generic Phase Diagram for $R_2\text{PdSi}_3$ ($R =$ Heavy Rare Earth)?

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Abstract. The magnetic phase diagrams of hexagonal $R_2\text{PdSi}_3$ compounds have been found to feature an apparent similar phase when the field is applied along the respective magnetic easy axes. To elucidate this, neutron diffraction on single crystalline Tb$_2\text{PdSi}_3$ and Ho$_2\text{PdSi}_3$ in magnetic fields was performed. These two compounds have different magnetic structures in zero field. In Tb$_2\text{PdSi}_3$ the propagation vector is parallel to the hexagonal axis while in Ho$_2\text{PdSi}_3$ the propagation is in the basal plane. In both compounds the magnetic moment direction is perpendicular to the propagation vector. The application of a magnetic field along the respective easy axes destroys the zero field magnetic structure and leads to a ferrimagnetic phase with magnetic intensity on the same positions for both compounds. These findings and their connections to the crystallographic (super-)structure are presented and discussed.

1. Introduction

The $R_2\text{PdSi}_3$ ($R =$ heavy rare earth) compounds crystallize in an AlB$_2$-derived hexagonal structure (P6/mmm). The magnetic rare-earth ions form layers which can be understood as triangular lattice with latent geometric frustration. These layers are intersected by layers of the non-magnetic Pd and Si atoms. The stacking of the layers is along the c-axis. Experimental evidence from single crystal neutron diffraction strongly suggests the presence of Pd-Si order at the B positions [1]. Additional structural reflections were found on (m/2, 0, n/8) and (m/2, m/2, n/8) positions with $n, m$ as integers. No intensity was found on (0, 0, 1/2), (1/2, 1/2, 0) and (1/2, 1/2, 1/2) positions which would be expected from earlier published lattice constants by Kotsanidis et al. [2]. A detailed study of the crystallographic (super-)structure will be published elsewhere [3]. Here, only pre-published facts which are crucial for the understanding of the magnetic structure will be mentioned. The Pd-Si order doubles the lattice constant $a$ of the primitive unit cell and the (super-)structure reflections can then be described by a propagation vector (0, 0, 1/8). In the following, we describe the structure in the picture of the primitive AlB$_2$ structure and refer to the Pd-Si order as crystallographic superstructure. A direct
consequence of the doubling is the occurrence of two inequivalent rare earth positions. One of the rare earth positions (Wyckoff layer 1a) keeps still the hexagonal on-site symmetry while the other position (Wyckoff layer 3f) has monocline symmetry.

2. Phase diagrams

Ac-susceptibility measurements on single crystals grown in the Institut für Festkörper- und Werkstoffforschung, Dresden [4, 5] were analyzed for features like maxima, steps and kinks [6]. The position of these features in \((H, T)\) space has been used to derive phase diagrams. Similar features for different parameters are connected with lines as is shown exemplarily in figure 1, the phase diagram of Gd\(_2\)PdSi\(_3\) with the field applied along the \((H, 0, 0)\)-direction. The low temperature (below \(T_N\)), low field (below \(H_{c1}\)) part of the phase diagram labelled AFM displays a variety of metamagnetic phases, also for other \(R_2\)PdSi\(_3\) compounds (in most compounds also a second transition temperature \(T_2\) is observed).

The FiM labelled phase in figure 1 is of interest since it has an intriguing behavior with the phase boundary increasing with higher fields towards higher temperatures. Similar phases are found in the phase diagrams for \(R = \text{Gd, Tb, Dy and Ho}\) when a field is applied along the magnetic easy direction. For the compounds with a small magneto-crystalline anisotropy like Ho\(_2\)PdSi\(_3\) and Gd\(_2\)PdSi\(_3\) this phase can be found also in the magnetic hard direction. Therefore, the existence of a generic phase diagram as schematically drawn in figure 2 is proposed for the \(R_2\)PdSi\(_3\) (\(R = \text{heavy rare earth}\)) compounds.

![Figure 1: Magnetic phase diagram of Gd\(_2\)PdSi\(_3\) derived from ac-susceptibility measurements. The critical magnetic field \(H_{c1}\) and the Néel temperature \(T_N\) are marked.](image1)

The FiM phase might be interpreted as a ferromagnetic induced state. However, the critical field \(H_{c2}\) at \(T = 2\) K above which the compound is in the saturated state is found at high fields (9.4 T for Gd\(_2\)PdSi\(_3\); larger than 13 T for Tb\(_2\)PdSi\(_3\) and Ho\(_2\)PdSi\(_3\)). Thus we are left with the question about the the nature of the FiM phase.

![Figure 2: Proposal for a generic phase diagram for \(R_2\)PdSi\(_3\) when a field is applied along the magnetic easy axis. For details see text.](image2)

3. Neutron scattering in applied magnetic fields for Ho\(_2\)PdSi\(_3\) and Tb\(_2\)PdSi\(_3\)

Neutron diffraction measurements were performed at the Helmholtz-Zentrum Berlin (HZB, formerly HMI) at the E2 diffractometer for Tb\(_2\)PdSi\(_3\) (\(T_N = 23.6\) K; \(H_{c1} = 3.8\) T) and at the Laboratoire Leon Brillouin (LLB, Saclay) at the 6T2 diffractometer for Ho\(_2\)PdSi\(_3\) (\(T_N = 7.7\) K; \(H_{c1} = 0.8\) T). Both experiments used a graphite monochromator with \(\lambda/2\) filters at a wavelength of about 2.4 Å. The two investigated compounds are especially suited for the evaluation of a generic phase since their magnetic structures in zero field are completely different.
In zero field the magnetic structure of Tb$_2$PdSi$_3$ can be described with a propagation vector $\tau_{\text{Tb,1}} = (0, 0, 1/16)$ and that of Ho$_2$PdSi$_3$ with a propagation vector $\tau_{\text{Ho,1}} = (1/7+\delta, -2\delta, 0)$. In both cases the propagation vector is perpendicular to the magnetic moment direction.

The zero field magnetic structure of Tb$_2$PdSi$_3$ is influenced by the crystallographic superstructure. Magnetic intensity is found not only for $G + \tau_{\text{Tb,1}}$ where $G$ is a reciprocal lattice vector but also for $G+(1/2, 1/2, 0)+\tau_{\text{Tb,1}}$ according to the description of the crystallographic superstructure with the doubled basal plane. When a magnetic field is applied in the basal plane (in the magnetic easy direction) the zero field magnetic structure is gradually replaced by a magnetic structure with a ferromagnetic component and with an antiferromagnetic component with $\tau_2 = (0, 0, 1/8)$. This vector equals the propagation vector for the crystallographic superstructure. We therefore observe in applied fields a strong increase of intensity on the positions of the superstructure reflections. In figure 3 scans along $Q = (1/2, 1/2, L)$ are shown for different temperatures and fields. The (nuclear) reflections due to the crystallographic superstructure are very weak and barely to be seen for the chosen scale (green curve). The magnetic reflections with $\tau_{\text{Tb,1}}$ of the zero field magnetic structure (blue curve) disappear for fields above $H_{c1}$. They are replaced by reflections with $\tau_2$ (red curve at the positions of the crystallographic superstructure). They exist in the field and temperature range as indicated for the FiM phase in figure 2. Below $H_{c1}$ and $T_N$ the magnetic phases with $\tau_{\text{Tb,1}}$ and $\tau_2$ coexist.

In Ho$_2$PdSi$_3$ the zero field magnetic structure with propagation vector $\tau_{\text{Ho,1}} = (1/7+\delta, -2\delta, 0)$ has no magnetic intensity along the $(1/2, 1/2, L)$ direction as shown in figure 4. Only (nuclear) reflections due to the crystallographic superstructure (green curve) are observed in zero field. However, applying a magnetic field larger than $H_{c1}$ (0.8 T) in the magnetic easy direction leads not only to the destruction of the zero field magnetic structure (not shown here) but to the appearance of magnetic intensity on the positions $Q = (1/2, 1/2, n/8)$ of the crystallographic superstructure. Again, these reflections exist in the field and temperature range as indicated for the FiM phase in figure 2.

4. Discussion and Conclusion

The application of a magnetic field along the easy magnetic directions in Tb$_2$PdSi$_3$ and Ho$_2$PdSi$_3$ leads for both compounds to a similar magnetic structure with a ferromagnetic component and an additional antiferromagnetic component. This magnetic structure can be identified with a magnetic phase named FiM in the magnetic phase diagram of these two compounds. Moreover, this phase has been identified in macroscopic measurements also for other $R_2$PdSi$_3$ compounds leading to the idea of a generic phase diagram. The FiM phase of other $R_2$PdSi$_3$ compounds still needs to be investigated.
more closely with neutrons to confirm if a similar magnetic structure connected to the FiM phase in \( R_2 \text{PdSi}_3 \) and \( \text{Ho}_2 \text{PdSi}_3 \) is also found there.

The nature of the strong connection between magnetic and crystallographic superstructure in \( R_2 \text{PdSi}_3 \) is open for speculation. In the following two possible interpretations are discussed briefly. The possibility of the FiM phase to be the ferromagnetic induced state is discarded since the absolute value of the magnetization is still below \( gJ \) in the FiM phase.

The first interpretation proposes a strong crystalline electric field (CEF) effect which is relevant when the magnetic structure due to the exchange interactions is destroyed by a magnetic field \( (> H_{c1}) \). Additionally, a pronounced difference of the CEF on the two different rare earth sites due to the crystallographic superstructure has to be assumed. The surrounding CEF defines the ground state wave function of the rare earth ion. Since the rare earth ions have different surroundings (either they are neighbored by Pd and Si or by Si alone) different ground states are imaginable. The FiM phase is then a true ferromagnetic phase but with different magnitude of the magnetic moments. The change of the magnetic intensity with the magnetic field can be explained by the changing of the mixing ratio of the wave functions. However, this model cannot explain the potential existence of a FiM phase for \( \text{Gd}_2 \text{PdSi}_3 \). In addition, evidence for different magnetic moments in zero field due to different ground states is missing [7].

The second interpretation on the other hand assumes a strong exchange interaction. Then the CEF only defines the magneto-crystalline anisotropy (moment direction). Though strong exchange interactions between neighboring rare earth ions are present at higher temperatures the Néel temperature is diminished due to geometric frustration of the rare earth ions. Furthermore, the effect of geometric frustration is confined to layers of the rare earth ions. The application of a magnetic field lifts the geometric frustration in the layer and nearest neighbor exchange becomes relevant. The nearest neighbor exchange is antiferromagnetic and yields an antiferromagnetic in plane component following the \( 2*a \) basal plane. The sequence of this antiferromagnetic component along the \( c \)-axis is modulated in the sequence of the Pd/Si layers. Then the crystallographic superstructure influences the exchange from rare earth layer to rare earth layer and consequently the resulting magnetic structure follows the modulation of the superstructure.

For a final interpretation of the nature of the similar magnetic structures found in \( R_2 \text{PdSi}_3 \) more measurements are needed. These are aimed to reveal the nature of the FiM phases in other \( R_2 \text{PdSi}_3 \) compounds. The strong coupling of the magnetic properties to the crystallographic (super-)structure and the possibility to influence this coupling via an external magnetic field in the \( R_2 \text{PdSi}_3 \) compounds make this series interesting for further studies.

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