Neutron diffraction study of magnetic structures in single crystal Ho$_2$PdSi$_3$ in magnetic fields up to 5 T

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Abstract.
Due to the interplay between RKKY exchange interaction, crystalline-electric field (CEF) effects and geometric frustration due to the AlB$_2$-derived hexagonal crystal structure, the study of R$_2$PdSi$_3$ ($R = $ rare earth) has been found to be a challenging field in rare earth magnetism. In this contribution we present the results of neutron diffraction experiments on a Ho$_2$PdSi$_3$ single crystal in the magnetically ordered state at $T \leq 1.6$ K. The compound orders antiferromagnetically in the basal plane with the propagation vector $\tau = (1/7-\delta, 2\delta, 0)$ where $\delta \sim 0.01$ r.l.u. In magnetic fields applied along the (110) magnetic hard axis, the structure persists up to the highest measured field of 5 T. The dependence of the intensities as a function of field suggests that the $\tau$ structure is a single-k multi-domain structure.

1. Introduction
The series of R$_2$PdSi$_3$ ($R = $ rare earth) compounds have 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$^2$. A rich variety of magnetic phenomena from long-range antiferromagnetic order with propagation vectors in different directions to short-range magnetic correlations have been observed in the series$^3$. This complex magnetic behavior results from the interplay between RKKY interaction, magneto-crystalline anisotropy based on crystal-electric field (CEF) effects and geometric frustration due to the AlB$_2$-derived hexagonal crystal structure.

A relatively weak magneto-crystalline anisotropy makes Ho$_2$PdSi$_3$ an interesting compound for the study of the delicate balance between the various interactions. The Stevens factor ($\alpha \sim -0.022$) for Ho$^{3+}$ suggests a magnetic easy axis in the basal plane which is also observed in high-temperature susceptibility data. However, in the magnetically ordered state the magnetic easy direction is the (001) direction. The exchange of the magnetic easy and hard axes is observed at around 50 K$^4$. The Néel temperature $T_N$ is 7.7 K, a second phase transition occurs at $T_2 = 2.3$ K$^4$. 

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Neutron diffraction measurements on powder Ho$_2$PdSi$_3$ samples were performed in zero field by Szytula et al.$^2$ proposing a magnetic structure with c-axis oriented, sine-wave modulated magnetic moments with a propagation vector of (0.137, 0.0066, 0) at $T = 1.5$ K. In the present paper we present the results of neutron diffraction measurements on a single crystalline Ho$_2$PdSi$_3$ sample in zero field and in fields up to 5 T applied in the direction of the magnetic hard axis (110).

2. Experimental details

The single crystal preparation and characterization are described elsewhere$^{[5, 6]}$. The neutron scattering experiments were carried out at the Helmholtz-Zentrum Berlin (formerly HahnMeitner Institut) with the E2 diffractometer and at Laboratoire Léon Brillouin on the 6T2 diffractometer with lifting counter option. The Ho$_2$PdSi$_3$ (m $\simeq$ 2.9 g) single crystal used in the studies is slightly irregular in shape with 6 oriented faces ground on a cylindrical sample of 9 mm length and 5 mm diameter. On both diffractometers a graphite monochromator was used with additional pyrolytic graphite filters to suppress $\lambda/2$ contaminations.

The E2 experiment used a standard orange-type cryostat capable of reaching 1.5 K base temperature. The E2 diffractometer employs a “banana”-type multidetector with 400 channels covering a $2\theta$ angle of 80$^\circ$. The sample is rotated around the axis perpendicular to the $HK0$ scattering plane in steps of 0.2$^\circ$. The used neutron wavelength was 2.39 Å.

For the 6T2 experiment, the sample was mounted in a $^4$He cryostat (base temperature $\sim$ 1.5 K) within a vertical magnet with the $(H, H, 0)$ direction parallel to the field direction making the $HH0$ plane the scattering plane. The diffractometer has a “classical” setup with a single detector mounted on a lifting arm to allow access to reflections off the scattering plane. It is worth noting that the $Q$ resolution along the perpendicular direction is not as good as along directions in the scattering plane (normally the FWHM is about two times larger). The used neutron wavelength was 2.345 Å. The sample position was not very accurate in height and therefore produced an offset of the zero position in the reciprocal space.

3. Results and Discussion

3.1. The magnetic structure of $\tau_{\pm} = (1/7\mp\delta, \pm 2\delta, 0)$

Figure 1 shows the full reciprocal $HK0$ plane of Ho$_2$PdSi$_3$ at $T = 1.5$ K obtained on E2. Magnetic Bragg peaks appear on positions {$G \pm r_0$} where $G$ is a reciprocal lattice vector and $r_0 = (1/7, 0, 0)$. Closer examinations show that the magnetic reflections are split in directions perpendicular to the “main” propagation direction $(H, 0, 0)$. Thus the actual propagation vectors are $\tau_{\pm} = (1/7\mp\delta, \pm 2\delta, 0)$. Shown in the upper left part of figure 2 is the centered $Q$ scan measured on 6T2 at $Q_0 = (0, 0, 1) + (1/7, -1/7, 0)$ where the splitting of $\tau_{\pm}$ is nicely resolved with $\delta \delta \sim 0.01$ r.l.u. in $(\pm \delta, \pm \delta, 0)$. At $Q_0 = (0, 0, 1) + (0, -1/7, 0)$ (upper right in figure 2) it is more difficult to resolve the splitting because the scan direction is along the 210 direction which is out of the scattering plane. The appearance of the higher harmonics at 3 $\tau_{\pm}$ and 5 $\tau_{\pm}$ in figure 1 indicates that the magnetic structure is not a simple sinusoidal-modulated structure. The temperature dependence of higher harmonics suggests that the squaring-up process is closely related to the transition at $T_2$ ($\sim$ 2 K), similar to the one observed in Er$_2$PdSi$_3$ compound$^3$.

3.2. Domain effect of the magnetic structure $\tau_{\pm} = (1/7\mp\delta, \pm 2\delta, 0)$

Due to the difficulty to resolve the two splits of $\tau_{\pm}$, only the summed intensity of $\tau_{+}$ and $\tau_{-}$ can be compared. They are labeled as $\tau^0_r$. With the 6T2 experiment, the field dependence of the $\tau^0_r$ intensities has been carefully followed at $T = 1.6$ K from $\mu_0H = 0$ T to 5 T. Transverse scans along five (of the six) symmetric equivalent vectors of $\tau_0$ that were accessible with the lifting counter were performed (see figure 3 for the relative orientations).
Figure 1. Full reciprocal HK0 plane of Ho$_2$PdSi$_3$ at $T$ = 1.5 K obtained on E2. The reciprocal lattice directions (100) and (010) are marked with arrows. The two rings originate from the scattering on polycrystalline Al from the sample holder and the cryostat.

Shown in figure 2 is a comparison between $\tau^0_0$ and $\tau^2_0$ at 0 T and 5 T, respectively. The intensity of the $\tau^0_0$ reflection exhibits a large increase in applied field while the $\tau^2_0$ reflection is already oppressed at 5 T. In figure 4 the field dependence of the integrated intensities of all measured $\tau^i_0$ is shown. The clear trend of the increase of the intensity of $\tau^0_0$ with the decrease of the other $\tau^i_0$s suggests that the magnetic structure is a multi-domain structure. And the fact that the $\tau^0_0$ intensity at 5 T is only slightly less than the sum of the $\tau^0_1, 2$ intensities at zero field indicates a domain repopulation.

The magnetic structure is rather robust for fields along the (1\bar{1}0) direction (persistent up to 5 T at least). This could be due to the fact that the antiferromagnetic couplings are between the magnetic components parallel to the $c$-axis. The application of a magnetic field perpendicular to the moment direction only cants the magnetic moment gradually.

Figure 2. Centered Q scans of Ho$_2$PdSi$_3$ measured on 6T2 at $T = 1.6$ K at $Q_0 = (0, 0, 1) + \tau^0_0, \tau^2_0$ where $\tau^0_0 = (1/7, -1/7, 0)$ (left frames) and $\tau^2_0 = (0, -1/7, 0)$ (right frames) in zero field (upper frames) and at 5 T (lower frames). The solid lines are the fitted values. For details see text.
4. Conclusions
The field dependence of the magnetic structures of Ho$_2$PdSi$_3$ has been studied using neutron diffraction measurements on a single crystalline sample. Zero field measurements revealed propagation vectors of $\tau_0^\pm = (1/7 \mp \delta, \pm 2\delta, 0)$ with $\delta \sim 0.01$ r.l.u. Furthermore, higher harmonics of $\tau_0^\pm$ have been observed below the Néel temperature suggesting a squaring-up of the magnetic moments at low temperature. This is consistent with the theoretical observation that the sinusoidal modulated structure is generally not a ground state[7]. The field dependent measurements of the $\tau_0^\pm$ structure show that it is a multi-domain structure. There are six domains, each related to one of the six symmetric equivalent directions of $\tau_0^\pm$. The application of an external field along one of the six $\tau_0^i$s (110 here) suppresses the domains with other propagation vectors, leaving only the domains with propagation vectors ($\tau_1^\pm$) most parallel to the field.

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