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Magnetic transitions in YbCo$_2$Si$_2$

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Abstract. The results of powder neutron diffraction studies of an antiferromagnet YbCo$_2$Si$_2$ with $T_{N,L}=0.9$ K and $T_{N,H}=1.7$ K were reported. An appearance of several superlattice peaks was observed at 0.48 K, which characterizes the antiferromagnetic ground state in YbCo$_2$Si$_2$. By heating up to 1.1 K, to the intermediate phase, a substantial change in the magnetic reflection from the ground state was revealed. The magnetic peak shifts to lower angle, indicating longer periodicity in the intermediate phase. In addition, the intensity ratio of the magnetic peaks shows significant change between two phases. In contrast to other magnetic RCo$_2$Si$_2$, the simple two-sublattice magnetic structure characterized by an existence of the magnetic 1 0 0 reflection cannot account for the observed magnetic peaks at both temperatures, which suggests more complex magnetic structures in YbCo$_2$Si$_2$.

1. Introduction
Magnetic order in ytterbium-based intermetallic compounds is not often realized. This mainly results from 4f character of ytterbium; the compound tends to exhibit valence fluctuation and/or strong coupling to conduction electron which leads to non-magnetic state or weak magnetism. YbCo$_2$Si$_2$ is one of examples in which an ytterbium ion is almost trivalent and hence shows magnetic ordering at $T=1.7$ K[1, 2, 3]. Recently, we have succeeded to grow high-quality large single crystals of YbCo$_2$Si$_2$. A detailed study using the single crystal allows us to clarify an additional first order transition at $T_{N,L}=0.9$ K below $T_{N,H}=1.7$ K in YbCo$_2$Si$_2$[4]. Furthermore, this compound was found to exhibit a complex magnetic phase diagram for small applied fields, lower than $B=2$ T[5]. The nature of these magnetic transitions are still unclear at present.

YbCo$_2$Si$_2$ has another intriguing aspects; this compound is iso electronic to YbRh$_2$Si$_2$ which is known to be one of the model compounds to study quantum critical point [6, 7]. YbRh$_2$Si$_2$ undergoes an antiferromagnetic transition at very low temperature, $T_N=70$ mK. An application of tiny magnetic fields of less than 1 T is sufficient to suppress the antiferromagnetic order and leads to the field-induced antiferromagnetic quantum critical point. On the other hand, the weak antiferromagnetism in YbRh$_2$Si$_2$ makes it difficult to study detailed magnetic properties, such as magnetic structure, in this compound. Obviously, the ordering temperature of YbCo$_2$Si$_2$ is much higher than that of YbRh$_2$Si$_2$. The Mössbauer spectroscopy and the magnetization
Figure 1. Neutron powder diffraction patterns of YbCo$_2$Si$_2$ measured at 0.48 K (the ground state), 1.1 K (the intermediate phase) and 2.0 K (the paramagnetic state). Arrows at 0.48 K and 1.1 K indicate magnetic peaks.

measurements indicate a large ordered moment exceeding 1 \( \mu_B \) in YbCo$_2$Si$_2$[1, 4]. Therefore, YbCo$_2$Si$_2$ is not only an interesting system in its own to study its magnetic properties but also a good reference to YbRh$_2$Si$_2$. In order to reveal the magnetic structure of YbCo$_2$Si$_2$, a neutron powder diffraction experiment was carried out.

2. Experiment
A polycrystalline sample of YbCo$_2$Si$_2$ was obtained by the flux method. The characterization by powder x-ray diffraction confirmed that the present sample consists of a single phase. Neutron powder diffraction experiments were carried out on the diffractometer E2 installed at the reactor hall of the BER-II reactor in the Helmutz-Zentrum Berlin, Germany. Neutrons with a wavelength of 2.4 Å were provided by the pyrolytic graphite monochromator. An incident
collimator of 30° and a radial oscillating collimator after the sample were used in order to achieve good signal-to-noise ratio. The YbCo$_2$Si$_2$ powder with a total mass of about 7 g was sealed in a Cu container together with small amounts of deuterized methanol-ethanol as refrigerant for good thermal contacts. A 3He cryostat was used for cooling the sample well below the lower transition temperature of $T_{N,L}=0.9$ K.

### 3. Results and Discussion

Figure 1 shows neutron powder diffraction patterns of YbCo$_2$Si$_2$ measured at 0.48 K ($T < T_{N,L}$, the ground state), 1.1 K ($T_{N,L} < T < T_{N,H}$, the intermediate phase) and 2.0 K ($T_{N,H} < T$, the paramagnetic state). The observed patterns at 2.0 K can be indexed by the reported structure of YbCo$_2$Si$_2$ with the space group $I4/mmm$. A strong peak at 70° and some broad structures in the background are attributed to Cu and the refrigeration medium, respectively.

By cooling down below 2.0 K, several superlattice reflections were clearly observed in addition to the nuclear Bragg peaks. At the lowest temperature of 0.48 K, the strong peak was found at 19.1° as indicated by a thick arrow in the figure. Distinct peaks were also observed around 41° and 45°. Since the intensity of the superlattice peak is stronger at lower angle, these peaks could be attributed to the magnetic ones. At higher temperature of 1.1 K, in the intermediate phase, superlattice peaks were still observed in the similar position to the ground state. The magnetic peak at around 19° is obvious whereas those around 40° becomes unclear at this temperature. The nuclear part of the diffraction patterns as well as the background are almost identical in the present temperature range.

In order to get a clear view of a magnetic part of diffraction pattern at 0.48 and 1.1 K,
the nuclear part and the background are subtracted by using the data at 2.0 K. Figure 2 shows difference diffraction patterns of YbCo$_2$Si$_2$ at 0.48 K and 1.1 K around (a) 15° $\sim$ 30° and (b) 37° $\sim$ 48°. The magnetic part of the diffraction pattern is successfully extracted by this subtraction. This subtraction makes it clear that there are magnetic peaks also at 28° at 0.48 K and 39° at 1.1 K, which are overlapped onto the 0 0 2 and 1 0 1 nuclear Bragg peaks, respectively. By heating up to the intermediate phase, a magnetic peak position shifts to lower angle : 19.1° at 0.48 K to 18.6° at 1.1 K. Concomitantly, the peak intensity exhibits substantial changes. It should be noted that the peak intensity around 19° in the intermediate phase is stronger than that in the ground state, whereas the other peaks become weaker through the transition. This results in a drastic change in the intensity ratio between the two phases; the peak intensity ratios at $\sim$19°, $\sim$27° and $\sim$40° are $I_{19°} : I_{27°} : I_{40°} = 1 : 0.59 : 0.59$ and $1 : 0.20 : 0.12$ at 0.48 K and 1.1 K, respectively. Furthermore, the peak around 46° becomes broader in the intermediate phase as compared to that in the ground state.

Hereafter, we try to figure out the nature of the magnetic transitions in YbCo$_2$Si$_2$ from the present result. The observed magnetic peaks in both phases cannot be indexed with the 1 0 0 and/or 0 0 1 which is expected to find at around 36.3° and 14.2°, respectively. The absence of the 1 0 0 and 0 0 1 magnetic reflections reflects that the magnetic structures in both ordered states are not the simple one, and should have a longer periodicity. It should be pointed out that the 1 0 0 magnetic peak was observed in all other magnetic RC$_2$Si$_2$ compound, i.e., the magnetic coupling is identical among RC$_2$Si$_2$ except $R$=Yb which could be represented as the $+ - + -$ sequence along the tetragonal c-axis. The orientation of an ordered moment is not unique among them: parallel to the tetragonal axis in $R$=Pr, Nd, Ho, Tb and Dy whereas normal to it in $R$=Er and Tm[8, 9]. Thus YbCo$_2$Si$_2$ is the exceptional case in this family.

The substantial changes in the magnetic peaks between the ground state and the intermediate phase were clearly observed in the present study. The increase of the magnetic peak intensity around 19° in the intermediate phase could not simply imply an increase of the ordered moment, since other magnetic peaks become weaker. An apparent change in the intensity ratio below and above $T_{N,L}$ suggests that this transition possibly involves a structural change such as moment re-orientation together with the variation in the periodicity. Furthermore, the broadening of the peak at 48° in the intermediate phase may indicate a peak splitting from the ground state, in other words, the lowering of the symmetry in the propagation vector.

The present results could not allow us to determine the exact magnetic propagation vector of both phases. In order to reveal the conclusive magnetic structure of YbCo$_2$Si$_2$, single crystal neutron diffraction experiments are currently in progress.

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