Long-time variation of magnetic structure in rare-earth intermetallic compounds

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Abstract. Long-time variations of magnetic structure have been studied by means of time-resolved neutron scattering measurements in two kinds of non-diluted uniform magnets, PrCo₂Si₂ and TbNi₂Si₂. These materials show commensurate (C) to incommensurate (IC) magnetic transitions below Néel temperature. When a sample is cooled to the low temperature C-phase, magnetic Bragg scattering signals from the C and the IC phases coexist and then their intensity vary with time. These characteristics are similar to those of the previously studied material CeIr₃Si₂. However, the temperature ranges in which clear time variations exist are much narrower in the present materials.

1. Introduction

Recently, long-time variation of magnetic structure has been observed in a non-diluted uniform magnet CeIr₃Si₂ [1]. This material shows successive magnetic transitions in zero magnetic field and multi-step metamagnetic transitions at relatively low magnetic field. These observations suggest that the frustrating magnetic interactions cause the long-time variation of magnetic structure in a material without random magnetic interactions. We have searched for other materials which show similar long-time variation of magnetic structure. We have found two intermetallic compounds PrCo₂Si₂ and TbNi₂Si₂.

Magnetic properties of intermetallic compounds REM₂X₂, where RE, M and X stand for a rare earth metal, a transition metal and silicon or germanium, respectively, have been extensively studied since 1980’s [2]. These materials crystallize in the body centered tetragonal structure (space group I₄/mmm), with RE, M and X atoms occupying the 2a, 4d and 4e positions, respectively. PrCo₂Si₂, TbNi₂Si₂ and some of other REM₂X₂ compounds show similar characteristics found in CeIr₃Si₂, namely the successive magnetic transitions and multi-step metamagnetic transitions at relatively low magnetic field.

Neutron diffraction study of PrCo₂Si₂ using a single crystal sample clarified the magnetic structure [3]. Three different magnetic phases were found below Tₘ=30 K. For all these phases, the magnetic moments of Pr atoms (∼3.2 µB at 4.2K) on the same (001) plane align ferromagnetically parallel or antiparallel to the c-axis. Therefore, the magnetic propagation vector is given as k = (0, 0, k) [2π/c]. For T < T₁=9 K, the sequence of ferromagnetic planes is +−+− which corresponds to the propagation vector of k=1 (C-phase). It transforms to the incommensurate structure with k=0.926 at T₁ (IC₁-phase). In this phase, the presence of the
third harmonics of Bragg peaks indicates a square-wave structure. Above $T_2=17$ K, another incommensurate structure with $k=0.777$ develops and persists up to $T_N$ (IC$_2$-phase). High-field magnetization measurements along the $c$-axis at 4.2 K showed four steps of increase at fields 1.2, 3.8, 6.7 and 12.2 T.

Neutron diffraction studies of TbNi$_2$Si$_2$ were made by several groups [2, 4, 5, 6]. It was shown that a simple antiferromagnetic structure with a $+-+$ sequence of ferromagnetic (110) planes appears below $T_t=7.9$ K (C-phase). Above $T_t$, an incommensurate structure with $k = (1/2 + \tau, 1/2 - \tau, 0)[2\pi/a]$, where $\tau = 0.074$ appears and persists up to $T_N$=14.6 K (IC$_2$-phase). The magnetization value measured at $T=1.3$ K with the field parallel to the $c$-axis jumps at five critical fields of 1.5, 2.2, 2.4, 4.4 and 4.6 T. Above 4.6 T it reaches the saturation value of 8.8 $\mu_B$/f.u., which is in good agreement with the results of the neutron experiments.

Based on these previous works, we made macroscopic and neutron scattering experiments to detect time variations of magnetic structure after rapid change of temperature across the magnetic transition temperatures. In this report, we present the results of time-resolved neutron scattering experiments of these two materials.

2. Experimental

Single crystal samples of PrCo$_2$Si$_2$ and TbNi$_2$Si$_2$ were cut from the ingots grown by the Czochralski pulling method and used in the previous works [3, 6]. Magnetization measurements detected no appreciable aging effect in these samples. The mass of each sample is 0.21 g and 0.34 g, respectively. Neutron scattering experiments of PrCo$_2$Si$_2$ were conducted using the 4G triple-axis spectrometer installed at the JRR-3M reactor of JAEA-Tokai. Measurements were made by a double-axis mode using pyrolytic graphite crystals for a monochromator and a filter. The wave-length of the incident neutron and the horizontal beam collimation are 2.35 $\AA$ and 40$\prime$ - 40$\prime$ - 40$\prime$, respectively. The sample was mounted with the [001] direction vertical and the magnetic Bragg peaks in the $a^*-c^*$ plane were measured.

Neutron scattering experiments of TbNi$_2$Si$_2$ were made using the T1-1 triple-axis spectrometer installed at the thermal guide of the JRR-3M reactor. Measurements were made using pyrolytic graphite crystals for a monochromator and a filter. The wave-length of the incident neutron and the horizontal beam collimation are 2.46 $\AA$ and guide - 40$\prime$ - 40$\prime$, respectively. The sample was mounted with the [001] direction vertical and the magnetic Bragg peaks in the $a^*-b^*$ plane were measured. A closed-cycle refrigerator was used for each experiment.

3. Results and discussion

Figure 1 shows the temperature variations of the amplitude of three kinds of magnetic Bragg diffractions. The measurements were made with heating and cooling processes. In each process, scans across the peaks were started after the sample temperature reached the target temperature same as usual measurements. The time required for the measurement at one temperature was typically 20 min. The result is basically same as the previous one [3], which showed the presence of three magnetic phases. In the present measurements, clear thermal hysteresis has been observed at the boundary between the C and IC$_2$ phases.

Figure 2 shows the time evolutions of neutron scattering patterns from (a) the C-phase and (b) the IC$_1$-phase of PrCo$_2$Si$_2$ at $T=10$ K after cooled from 11 K. The amplitude of the C-phase signal increases with time whereas the amplitude of the IC$_1$-phase signal decreases. Neither the center position nor the width of each signal changes with time.

Time variation measurements of these signals were made at various temperatures. Figure 3 shows the results at representative temperatures. Clear time variations of magnetic scattering intensity exist in the narrow temperature range around $T_1$. The time variations of the signal amplitude can not be expressed by a simple exponential function. We tentatively analyzed the
Figure 1. Temperature variations of the amplitude of three kinds of magnetic Bragg diffractions of PrCo$_2$Si$_2$.

Figure 2. Time evolutions of neutron scattering patterns from (a) the C-phase and (b) the IC$_1$-phase of PrCo$_2$Si$_2$ measured at 10 K after cooled from 11 K. Curves represent the results of the least squares fittings described in the text.

observed time variation as a sum of two exponential functions having different characteristic times. The curves in Fig. 3 show the results of the fittings.

Figure 3. Time variations of the amplitude of neutron scattering signals from (a) the C-phase and (b) the IC$_1$-phase of PrCo$_2$Si$_2$ at various temperatures. Curves represent the results of the fittings described in the text.

Figure 4 shows the temperature variations of the amplitude of two kinds of magnetic Bragg diffraction signals from TbNi$_2$Si$_2$. New features of the temperature variation behavior have been observed in addition to the previous result [6]. Firstly, large thermal hysteresis has been observed in wide temperature region. Secondly, the amplitude of the C-phase signal at low temperature varies with the cooling speed. Based on these observations, we made time variation measurements of neutron diffraction signals.

Figure 5 shows the time variations of the amplitudes of (a) the C-phase and (b) the IC-phase signals at representative temperatures. The observed time variation behavior is basically same as the case of PrCo$_2$Si$_2$. 
Figure 4. Temperature variations of the amplitude of two kinds of magnetic Bragg diffractions of TbNi$_2$Si$_2$.

Figure 5. Time variations of the amplitude of neutron scattering signals from (a) the C-phase and (b) the IC-phase of TbNi$_2$Si$_2$ at various temperatures.

We have shown that magnetic Bragg peaks corresponding to different types of magnetic structures coexist near the transition temperatures and the amplitudes of these peaks vary with time after the change of sample temperature in both materials. We note that only the amplitude of Bragg peaks vary with time. Neither the position nor the line width showed appreciable time variation. These results indicate that we have observed the change of the volume fractions of two different magnetic phases same as the case of previously studied material CeIr$_3$Si$_2$ [1]. It should be noted that in the present materials time variations have been observed only in narrow temperature range around the commensurate to incommensurate transition temperatures.

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