Long-time variation of magnetic structure in CeIr$_3$Si$_2$

Kiyoiichiro Motoya$^1$, Yuji Muro$^1$ and Toshiro Takabatake$^2$

$^1$Department of Physics, Faculty of Science and Technology, Tokyo University of Science, Noda 278-8510, Japan
$^2$Department of Quantum Matter, ADSM, Hiroshima University, Higashi-Hiroshima 739-8530, Japan

E-mail: motoya@ph.noda.tus.ac.jp

Abstract. CeIr$_3$Si$_2$ shows successive magnetic transitions at $T_{N1}=4.1$ K and $T_{N2}=3.3$ K. At $T < T_{N2}$ it shows three-step metamagnetic transitions below $H=1.43$ T. In this non-diluted compound a long-time variation of magnetic structure has been detected by means of magnetic susceptibility and time-resolved neutron scattering measurements. When a sample is rapidly cooled below $T_{N2}$, the magnetic Bragg peaks corresponding to the intermediate temperature phase ($T_{N2} < T < T_{N1}$) are observed. The amplitude of these Bragg peaks gradually decreases with time. On the other hand, another group of Bragg peaks corresponding to the low temperature phase ($T < T_{N2}$) gradually grow with time. The characteristic time for these variations follows the Arrhenius law with an activation energy $E_a/k_B=4$ K. This is the first observation of long-time variation of magnetic structure in non-diluted uniform magnets.

1. Introduction

In the spin-glass study, long-time variations of magnetic properties have been extensively investigated. These properties have been regarded as characteristic behavior due to the multi-valley structure of the free energy which comes from random magnetic interactions. Therefore, in a system without randomness or imperfections we have not expected to observe time variations of magnetic properties in an attainable time scale. Recently, we observed a long-time variation of the magnetic structure in a non-diluted uniform magnet CeIr$_3$Si$_2$.

The first report of the magnetic property of this material was presented by Umarji and coworkers in 1987 [1]. They reported that the crystal structure of CeIr$_3$Si$_2$ was the CeCo$_3$B$_2$-type and that no magnetic transition had been observed down to 4.2 K. The magnetic and transport properties of CeIr$_3$Si$_2$ have been reinvestigated with polycrystalline sample by Muro and coworkers [2]. They found that CeIr$_3$Si$_2$ crystallizes not in the hexagonal CeCo$_3$B$_2$-type, but in the orthorhombic ErRh$_3$Si$_2$-type (Space Group Imma, No.74) that is a deformed structure of the CeCo$_3$B$_2$-type. Magnetic susceptibility and specific heat measurements revealed the existence of magnetic transitions at $T_{N1}=4.1$ K and $T_{N2}=3.3$ K. The isothermal magnetization curve at 1.9 K showed three-step metamagnetic transitions below $H=1.43$ T. Shigetoh and coworkers made magnetization, electrical resistivity and specific heat measurements utilizing a single crystal sample [3]. These measurements revealed easy-plane type anisotropic properties. The magnetization curves measured at $T=0.3$ K with a magnetic field along the $b$ or $c$ axis include a ferromagnetic component of 0.2-0.3 $\mu_B$/Ce and show metamagnetic transitions. Two metamagnetic transitions were observed at $H=0.68$ T and 1.3 T when the magnetic field was applied along the $b$-axis.
We started a neutron scattering study of CeIr$_3$Si$_2$ to clarify the magnetic structures in various magnetic phases and unexpectedly found long-time variations of the amplitudes of magnetic Bragg peaks. In this report we present the results of time variation of magnetic susceptibility and time-resolved neutron scattering measurements. We believe this is the first real-time observation of the magnetic structural change in a uniform magnetic system.

2. Experimental
A single crystal of CeIr$_3$Si$_2$ was grown by the Czochralski pulling method. Details of the procedure have been described in ref. 3. The lattice parameters at room temperature are $a = 7.178 \text{ Å}$, $b = 9.726 \text{ Å}$ and $c = 5.597 \text{ Å}$. The mosaic spread is $\sim 0.3^\circ$ (full width at half maximum, FWHM). For the magnetization measurements a small piece of single crystal (19 mg) was used. For the neutron scattering measurements a small piece ($\sim 2 \text{ mm} \times \sim 3 \text{ mm} \times \sim 3 \text{ mm}$) was cut from the ingot because of large neutron absorption of Ir atoms.

Magnetic susceptibility measurements were made using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design model MPMS). A magnetic field $H$ was applied parallel to the $b$-axis of the single crystal sample. Measurements were made with zero field cool (ZFC) and field cool (FC) processes. For each measurement, the sample was cooled from $T = 30 \text{ K}$ to the target temperature. A typical time needed to reach the target temperature was $\sim 7 \text{ min}$. Then the magnetic field was applied (for the ZFC process) and the magnetic susceptibility was repeatedly measured as a function of time $t$.

Neutron scattering experiments were conducted using the 4G and 5G triple-axis spectrometers installed at the JRR-3M reactor of JAEA-Tokai. The wave-length of the incident neutron and the horizontal beam collimation are $1.638 \text{ Å}$ and $40^\circ - 40^\circ - 40^\circ$, respectively. The sample was mounted with the [100] direction vertical in a closed-cycle $^3\text{He}$ gas refrigerator or a liquid $^4\text{He}$ cryostat. Time variations of neutron scattering patterns were measured after the sample was rapidly cooled from $T = 30 \text{ K}$ up to the target temperature at zero external field. The time required to cool the sample was $\sim 15 \text{ min}$.

3. Results and discussion
Time variations of magnetic susceptibility $M/H$ were measured with various combinations of $T$ and $H$. Figure 1 shows some examples of the results measured at $H = 50 \text{ Oe}$. In the low temperature (LT) phase below $T_{N2}$, the $M/H$ value measured with ZFC process increases rapidly after the application of $H$ at $t=0$. The increase continued up to the end of the measurements at $t \sim 23 \text{ h}$. On the other hand in the intermediate temperature (IT) phase ($T_{N2} < T < T_{N1}$), no appreciable time variation of $M/H$ was observed. The time variation at $T=1.9 \text{ K}$ after cooled in $H=50 \text{ Oe}$ is also shown. The $M/H$ value did not change appreciably until the magnetic field was turned off. When the magnetic field was turned off at $t=150 \text{ min}$, the magnetization value abruptly decreased and then the slow decrease continued.

Preliminary neutron scattering measurements showed that in the IT phase the magnetic Bragg peaks were observed at the $(0, 1.375, 0.64)$ and equivalent reciprocal lattice points (IT phase signal). On cooling the sample to the LT phase, these Bragg peaks decreased with time and another group of Bragg peaks appeared at the $(0, 4/3, 2/3)$ and equivalent points (LT phase signal) and these peaks grew with time. These observations have shown that the magnetic structure of the IT phase is maintained in the LT phase and it gradually transforms to the LT phase structure. We measured the time variations by repeating two types of scans along the $(0, \xi, \xi/2)$ direction across the $(0, 1.375, 0.64)$ and $(0, 4/3, 2/3)$ points alternately as illustrated in Fig. 2. The measurements were made at various temperatures for $\sim 24 \text{ h}$.

Figure 3 shows the neutron scattering patterns of the (a) IT and (b) LT phase signals taken at representative elapse times $t$ after cooled to $T=1.0 \text{ K}$. The amplitude of the IT phase signal decreases with increasing $t$. The amplitude of the LT phase signal increases with increasing $t$. 

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2
2.5
2.0
1.5
1.0
0.5
0.0 M / H (x10^-2 emu/g)

Figure 1. Time variations of magnetic susceptibility measured at various temperatures.

Figure 2. A illustration of two types of scans across the Bragg points of the IT and LT phase signals.

Figure 3. Time variations of neutron scattering patterns of the (a) IT and (b) LT phase signals measured at T=1.0 K. Curves represent the results of the least squares fitting.

For both signals no time variation of the peak position was detected. The line shape of the scattering patterns fits a Gaussian function of the instrumental resolution. When the sample was cooled to the IT phase, the IT phase signal immediately appeared and no time variation was detected.

Figure 4 shows the time variations of the amplitude of the (a) IT and (b) LT phase signals measured at various temperatures. At T=0.7 K the IT-phase signal was observed without detectable change of amplitude up to 10 h and no LT phase signal was observed. The time variations of the signal amplitude are well expressed by simple exponential functions posted in the figure. The characteristic time $t^*$ thus obtained are posted in the figure.

We analyzed the results in terms of the Arrhenius model. Figure 5 shows the Arrhenius plot of $t^*$ (log(1/$t^*$) vs 1/T plot). Temperature variation of 1/$t^*$ follows the Arrhenius law as shown by the straight line. The activation energy $E_a$ has been determined as $E_a/k_B = 4$ K.

We have shown the long-time variation behavior observed in the magnetis susceptibility and neutron scattering measurements. These results have shown that the magnetic structure of
Figure 4. Time variations of the amplitude of the (a) IT and (b) LT phase signals at various temperatures. Curves represent the results of the least squares fitting.

the IT phase is maintained below $T_{N2}$ and it gradually transforms to the LT phase structure. Although the magnetic structure in each phase has not been determined, we presume that the transition from the IT to LT phase is basically an incommensurate to a commensurate phase transition. We note that in the present measurements, only the amplitude of Bragg peaks varied with time. Neither the position nor the line width of Bragg peaks showed appreciable time variation. These results strongly suggest that we have observed the change of the volume fractions of two distinct magnetic regions with a commensurate and an incommensurate structures. The activation energy $E_a$ deduced from the Arrhenius law corresponds to the energy barrier accompanied by the motion of the interface between two regions. In future experiments with high intensity apparatus, we hope to detect the change of domain size at the early stage of the time evolution.

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