Real-time observation of magnetic structural change in the multistep metamagnet CeIr$_3$Si$_2$

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Abstract. A ternary intermetallic compound 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 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.

1. Introduction

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

The first report of the magnetic property of this material was presented by Umarji and coworkers in 1987 [1]. In this work they reported the crystal structure of CeIr$_3$Si$_2$ as the CeCo$_3$B$_2$-type and no magnetic transition was observed down to 4.2 K. Recently the magnetic and transport properties of CeIr$_3$Si$_2$ were 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. A 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. The Curie-Weiss behavior of the magnetic susceptibility curve between 100 K and 300 K shows a paramagnetic Curie temperature $\theta_p = -32$ K and an effective magnetic moment $\mu_{eff}=2.57$ $\mu_B$, close to that expected from a Ce$^{3+}$ free ion. The magnetic part of the electrical resistivity shows a maximum at $T_{max} \sim 15$ K accompanying a $-\ln T$ behavior between 20 and 50K. This shows that CeIr$_3$Si$_2$ is classified as a Kondo-lattice compound. Shigetoh and coworkers made magnetization, electrical resistivity and
specific heat measurements under various magnetic field utilizing a single crystal sample \[3\]. These measurements revealed the 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. When the magnetic was field applied along the $c$-axis a transition at $H=0.75$ T was followed by a linear increase of magnetization up to a kink at $H=3$ T. These values of critical magnetic field agree with those measured with a polycrystalline sample \[2\].

We started a neutron scattering study of CeIr$_3$Si$_2$ to clarify the magnetic structures in various magnetic phases. During this study we found long-time variations of the amplitudes of magnetic Bragg peaks. In this report we present the results of time-resolved neutron scattering measurements in zero external field. 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 has been described in ref. 3. For the neutron scattering measurements a small piece (\(\sim 2\) mm \(\times \sim 3\) mm \(\times \sim 3\) mm) was cut from the ingot because of large neutron absorption of Ir atoms. The lattice parameters at room temperature are $a=7.178$ Å, $b=9.726$ Å and $c=5.597$ Å. The mosaic spread is \(\sim 0.3^\circ\) (full width at half maximum, FWHM).

Neutron scattering experiments were conducted using the 4G and 5G triple-axis spectrometers 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 1.638 Å and 40’ - 40’ - 40’, respectively. The sample was mounted with the [100] direction vertical in a closed-cycle $^3$He gas refrigerator or a liquid $^4$He cryostat. Time variations of neutron scattering patterns were measured after the sample was rapidly cooled from $T=30$ K up to the target temperature at zero external field. The time required to cool the sample was \(\sim 15\) min.

3. Results and discussion
Preliminary measurements showed that in the intermediate temperature (IT) phase ($T_{N2}<T<T_{N1}$), magnetic Bragg peaks were observed at the (0, 1.375, 0.64) and equivalent reciprocal lattice points (IT phase signal). On cooling the sample below $T_{N2}$ to the low temperature (LT) phase, these Bragg peaks decreased with time. Concomitantly, another group of Bragg peaks appeared and grew with time at the (0, 4/3, 2/3) and equivalent points (LT phase signal). These observations have shown that the magnetic structure of the IT phase is maintained in the rapidly cooled LT phase and it gradually transforms to the LT phase structure.

In the present study, we have observed the time variations by repeating two types of scans across the (0, 1.375, 0.64) and (0, 4/3, 2/3) points alternately for \(\sim 24\) h at various temperatures. The measurements were made with scanning the spectrometer along the (0, $\xi$, $\xi/2$) direction to resolve two kinds of peaks.

Figure 1 shows the scattering patterns around (0, 1.375, 0.64) taken at representative elapse times $t$ after cooled to $T=1.9$ K. The amplitude decreases with increasing $t$ and no appreciable signal was observed for $t > 500$ min. Figure 2 shows the scattering patterns around (0, 4/3, 2/3) taken at representative $t$ at $T=1.9$ K. The amplitude of the signal increases with increasing $t$. For both of these measurements, no shift of the peak position with time was detected. The line shape of the scattering patterns fits a Gaussian function of the instrumental resolution.

Similar time variation measurements of the IT and LT phase signals were made at various temperatures. When the sample was cooled to the IT phase, the IT phase signal immediately appeared and no time variation was detected. Figures 3 and 4 show the time variations of
Figure 1. Time variation of neutron scattering patterns observed at $T=1.9$ K around the $(0, 1.375, 0.64)$ reciprocal lattice point.

Figure 2. Time variation of neutron scattering patterns observed at $T=1.9$ K around the $(0, 4/3, 2/3)$ reciprocal lattice point.

Figure 3. Time variations of the amplitude of the IT phase signal at various temperatures. Curves represent the results of the least squares fitting described in the text.

Figure 4. Time variations of the amplitude of the LT phase signal at various temperatures. Curves represent the results of the least squares fitting described in the text.

the IT and LT phase signals measured at various temperatures below $T_{N2}$. At $T=0.7$ K the IT-phase signal was observed without detectable change up to 10 h and no LT phase signal was observed. The time variations of the signal amplitude $A(t)$ are well expressed by simple exponential functions

$$A(t) = A_0 \exp\left(-\frac{t}{t^*}\right) \quad \text{and} \quad A(t) = A_0 \left[1 - \exp\left(-\frac{t}{t^*}\right)\right]$$

for the IT and LT phase signals, respectively. In these equations $A_0$ and $t^*$ are the constant value and the characteristic time for the amplitude variation, respectively. Curves in Figs. 3 and 4 show the results of the least squares fitting. For the LT phase signal, the asymptotic value decreases with increasing temperature. It is probably due to the normal temperature variation of the sub-lattice magnetization. Obtained values of $t^*$ are posted in the figures. $t^*$ values for the IT and LT phase signals are close for the same temperature.
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). $1/t^*$ follows the Arrhenius law as shown by the straight guide line in the figure. The activation energy $E_a$ of the time variation is $E_a/k_B=4$ K.

![Figure 5. Arrhenius plot of $1/t^*$ derived from the time variation of neutron scattering amplitudes of two kinds of signals.](image)

We have shown that two groups of magnetic Bragg peaks corresponding to different types of magnetic structure coexist in rapidly cooled CeIr$_3$Si$_2$. The amplitude of the Bragg peaks corresponding to the IT phase structure decreases and that of the LT phase increases with time. These results show that the magnetic structure of 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 phase to the LT phase is basically an incommensurate to 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 with the motion of the interface between two regions. In future experiments with high intensity apparatus, we will be able to observe the change of domain size at the early stage of the time evolution.

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