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Magnetic lock-in phase transition in $Tb_{0.95}Er_{0.05}Ni_5$ driven by low magnetic fields

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ABSTRACT

The magnetic properties of a mixed inter-metallic compound, $Tb_{0.95}Er_{0.05}Ni_5$, were investigated using a neutron diffraction method at low temperatures. These compounds were known to have a successive magnetic phase transition from the paramagnetic state at high temperature to a lock-in phase at low temperature through intermediate phases, i.e., PM(paramagnetic)–FM(ferromagnetic)–IC(incommensurate)–L(lock-in) in reverse order of temperature. A meta-magnetic phase transition between an IC phase and a FM phase at 9 K was observed with the critical field, $H_{MT}\sim 200$ mT. A new magnetic phase between the new phase (lock-in phase) and an IC phase has been observed. From the field dependence of the Bragg reflections and their satellite peaks at low temperatures (3–12 K), *weak field* driven first-order magnetic phase transitions were recorded at six fixed temperatures. The critical magnetic field decreases exponentially with the temperature. From these experimental results, we obtained a magnetic phase diagram of $Tb_{0.95}Er_{0.05}Ni_5$ at a low temperature region for the first time.
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1. Introduction

The compound, RNi_5 (where, R is a rare earth element), which shows a large magnetocrystalline anisotropy, crystallizes in a hexagonal $CaCu_5$ type structure [1]. The distinctive feature of RNi_5 is that the anisotropy energy of the R ions caused by the crystalline electric field exceeds the exchange energy by an order of magnitude [2,3]. This feature should lead to peculiar properties in the metamagnetic [illegible]

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paramagnetic RNi_5 and ferromagnetic $ErNi_5$ compounds [4–7]. Recently, it has been shown that $TbNi_5$ and $Tb_{0.925}Er_{0.075}Ni_5$ have FAN-like incommensurate magnetic ordering of Tb-ion magnetic moments described by two wave vectors ($\boldsymbol{k}_1=0$ and $\boldsymbol{k}_2=2\pi/c(0,0,\tau)$) [8,9]. The Tb-ion moments lie in the base plane and have mutually collinear ferromagnetic and modulated components [10]; however, the Er-ion moments are not ordered. Therefore, these intermetallics belong to the group of X–Y magnetic systems [11]. When an external magnetic field was applied to the $TbNi_5$ single crystal, the modulated component has vanished at about 350 mT and only the $\boldsymbol{k}_1$ vector (the ferromagnetic component) exists [12–14], i.e., the crystal undergoes a field induced incommensurate–commensurate (ICM–CM) magnetic phase transition. Taking into account the large magnetic anisotropy of RNi_5 compounds, it can be expected that the ICM–CM transition should be accompanied by the MT transition as well. However, because of overlaps of satellites with Bragg peaks (owing to the small magnitude of τ), the MT in the $TbNi_5$ could not be clearly observed.

In this paper, we report the observation of the MT in the [illegible] case compared to the case of $x=0$. This allows the assertion of the field dependence of τ and the magnetic moment (both the modulated and ferromagnetic components).

2. Materials and methods

A polycrystalline sample of $Tb_{0.95}Er_{0.05}Ni_5$ was prepared by an *induction melting method* using an alumina crucible under an argon atmosphere. According to the metallographic and x-ray analysis, it was found that the samples were synthesized as a single-phase $CaCu_5$-type structure after annealing at 1100 °C for 22 h under a pure helium atmosphere followed by quenching in cold water. A neutron powder diffraction experiment with an HRPD instrument at the HANARO reactor ($\lambda=1.8342$ Å) has been carried out. Vertical fields were applied to the sample using a Helmholtz coil magnet (0–52 mT, combined with a 4 K-CCR) and

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an electromagnet (0–800 mT, combined with a 10 K-CCR). Neutron powder diffraction patterns were analyzed using a refinement program, FullProf Suite [15].

3. Results and discussion

Fig. 1 shows parts of the neutron diffraction patterns of $Tb_{0.95}Er_{0.05}Ni_5$ at 3 K (a) and 30 K (b). When the temperature decreases, the intensities of the Bragg reflections (especially, (1 0 0), (1 0 1), (2 1 0) and their satellites) increase distinctly owing to the contribution of the magnetic scattering. From an analysis the neutron diffraction patterns, measured in a temperature range of 3–30 K, it can be concluded that the magnetic structure of $Tb_{0.95}Er_{0.05}Ni_5$ is a FAN-like one, which is similar to the case of $TbNi_5$ with wave vectors $\boldsymbol{k}_1=(0,\ 0,\ 0)$ and $\boldsymbol{k}_2=2\pi/c(0,\ 0,\ \tau)$, where $\tau=0.036$ at 20 K.

As shown in Fig. 1(c), the magnitude of τ decreases with temperature from 0.036 (at 20 K) to 0.027 (at 3 K), which can be expressed as a commensurate one with τ as 1/37, at 10 K, and it remains constant during a further cooling down to 3 K. As the sample is heated, τ increases, and there is a clear hysteresis in τ between the heating and cooling processes. As a result, the phase transition between the incommensurate phase and lock-in phase occurs at ~10 K. Note that the magnitude of τ for the $Tb_{0.95}Er_{0.05}Ni_5$ is 1.4-times larger than that for $TbNi_5$ [9].

When the external magnetic fields were applied to the sample at 9 K as shown in Fig. 2, the intensities of the satellite peaks decreased monotonically with the field, and diminished at fields higher than ~200 mT (a). On the other hand, the intensity of the Bragg reflection (central peak) increases visibly with a field up to H_{MT}≈200 mT, and then increases slowly. Analyzing the full diffraction patterns by Rietveld refinement, the magnetic moments of the ferromagnetic and the modulated components were obtained at each applied magnetic field. The ferromagnetic component increases with the applied field, but on the other hand, the modulated component decreases with it up to 200 mT, as shown in Fig. 2(b). Both components keep their value almost constant when the applied field exceeds 200 mT up to the maximum applied field of 800 mT. Therefore, the high field-induced ICM–CM (FM) phase transition occurs at H_{MT}≈200 mT at 9 K in $Tb_{0.95}Er_{0.05}Ni_5$, which is quite lower than $TbNi_5$, H_{MT}~350 mT at 2 K [14]. When the external field was turned off after reaching the maximum field, 800 mT, the magnetic moments did not go back to their initial states owing to the high magnetic anisotropy energy of the system.

Interestingly, another characteristic transition was found with an abrupt behavior of satellites and Bragg peaks at low field regions. Fig. 3 shows parts of neutron diffraction patterns near the (1 0 1) reflection. As one can see from the inset in Fig. 3, at 3 K, the intensities of the (1 0 1) Bragg peak and $(1\ 0\ 1)^{\pm}$ satellites do not vary with a field up to 42 mT. However, a diversity of intensities is clearly seen when the magnitude of the field exceeds the critical value of H_l≈43 mT. We observed the same phenomenon, a low field induced magnetic phase transition in $Tb_{0.925}Er_{0.075}Ni_5$ (H_l≈9 ± 1 mT at 4 K) when a similar experiment was carried out to confirm this hypothesis. In Fig. 4, according to temperature, the field dependences of intensity for ferromagnetic and modulated components show that the critical fields of the abrupt and anomalous magnetic phase transition decreased (43 → 15 → 12 → 7 mT) with an increase in temperature (3 → 5 → 6 → 9 K), respectively, and disappeared at 12 K. When the external magnetic field is turned off, after applying an external field up to 52 mT (above H_l=43 mT), the magnetic moments (both the FM and ICM

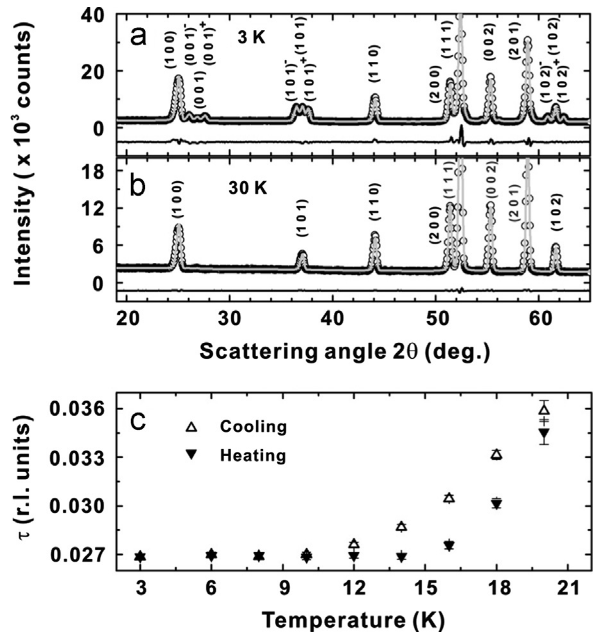

Fig. 1. The neutron diffraction diagrams of $Tb_{0.95}Er_{0.05}Ni_5$ measured at (a) 3 K and (b) 30 K (open circles; observed intensities, line; calculated profile, bottom line; difference between the experimental and calculated intensity), and (c) the temperature dependence of the wave vector $\mathbf{k_2}=2\pi/c(0,\ 0,\ \tau)$.

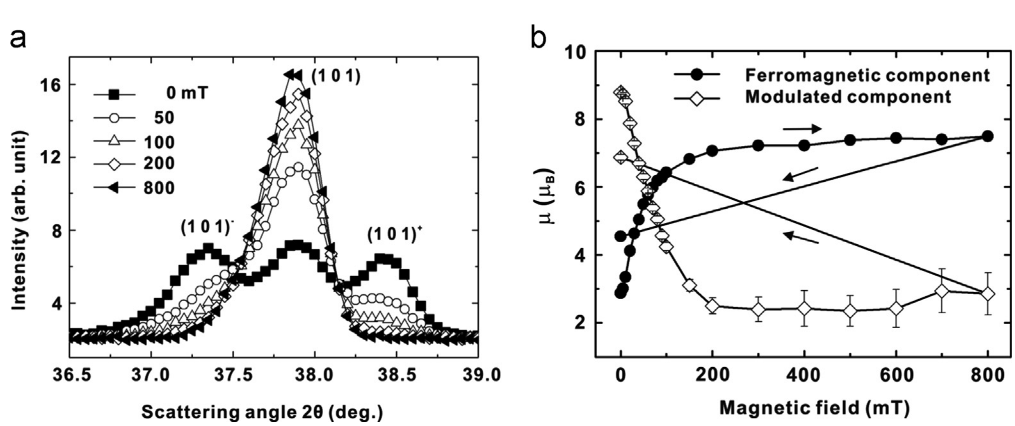

Fig. 2. (a) Evolution of neutron diffraction patterns around (1 0 1) reflection according to the applied magnetic field at 9 K, and (b) the magnetic field dependence of magnetic moments obtained by Rietveld refinement at each point. The arrows indicate the sequence of measurements. When the applied field is suddenly switched off, the magnetic moments do not return to their initial values.

components) kept their last condition at low temperature (the arrows in the figures). On the other hand, it returns to the initial state ($H=0$) with a growing temperature. From this result, it can be estimated that the energy for keeping the magnetic state competes with the thermal energy of the system at low temperature.

In summary, a neutron diffraction experiment on the $Tb_{0.95}Er_{0.05}Ni_5$ compound was carried out to study the magnetic behaviors at various temperatures and external magnetic fields. A metamagnetic transition, where the ICM structure slowly transforms into the CM (FM) structure according to the increasing magnetic field, was observed at $H_{MT}\sim$ 200 mT (at $T=9$ K). Through a careful investigation of a weak field effect on the diffraction patterns of the $Tb_{0.95}Er_{0.05}Ni_5$, a new magnetic phase transition (lock-in phase–ICM) was observed at a low temperature region for the first time. Recently, we have reported the lock-in phase transition in a $TbNi_5$ single crystal at a zero magnetic field, $T_l\sim$ 10 K [10]. In this study, the phase diagram of a similar compound, $Tb_{0.95}Er_{0.05}Ni_5$, between the lock-in phase and the ICM phase is deduced in H-T space by systematic isotherm measurements at several temperatures, 3–12 K. After the transition from the lock-in phase into the ICM by external field, it did not go back to the initial state when the applied field was suddenly turned off, and resembled a metamagnetic transition (ICM–FM).

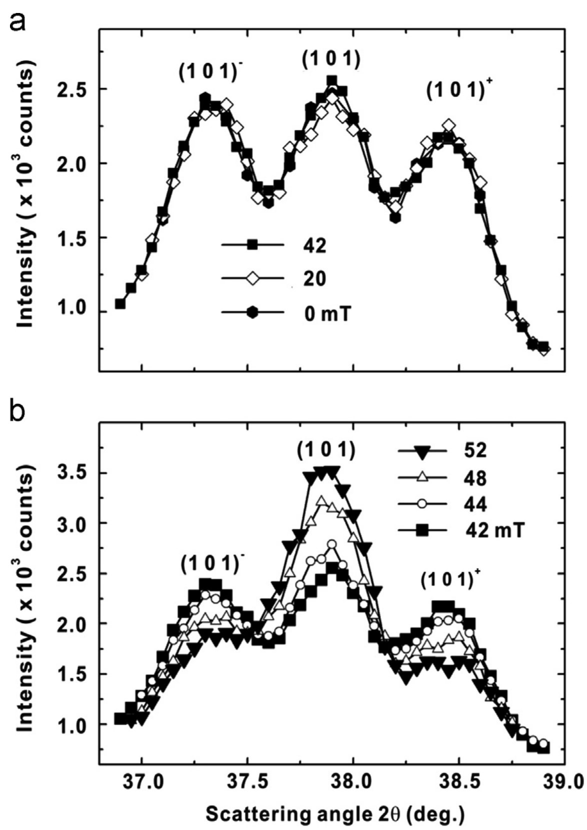

Fig. 3. A new magnetic phase transition driven by a weak magnetic field at 3 K. (a) Nearly no change up to 42 mT was observed but (b) an abrupt change between 42 mT and 44 mT has been observed.

4. Conclusion

According to the experimental results, an approximate phase diagram of $Tb_{0.95}Er_{0.05}Ni_5$ was depicted in Fig. 5. The magnitude of H_l in the $Tb_{0.95}Er_{0.05}Ni_5$ compound, as displayed at the bottom of Fig. 5, decreases drastically as the temperature increases and

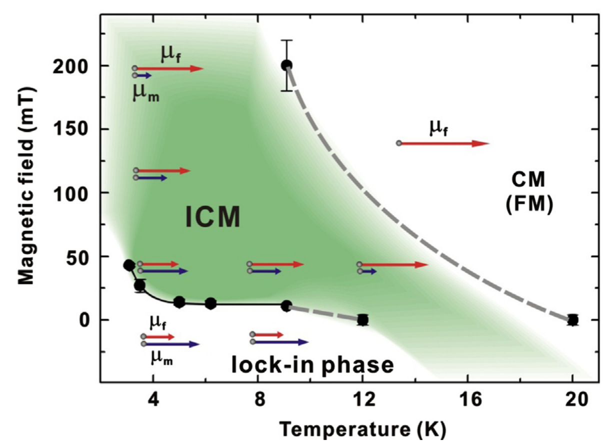

Fig. 5. A magnetic phase diagram of the $Tb_{0.95}Er_{0.05}Ni_5$. The solid line was fitted by an exponential equation from the temperature dependence of the critical field (H_l). The dashed lines are possible phase boundaries between the adjacent magnetic phases. Arrows indicate the spin alignment of the $Tb_{0.95}Er_{0.05}Ni_5$ for a phase transition relevant to the given temperature and external magnetic field.

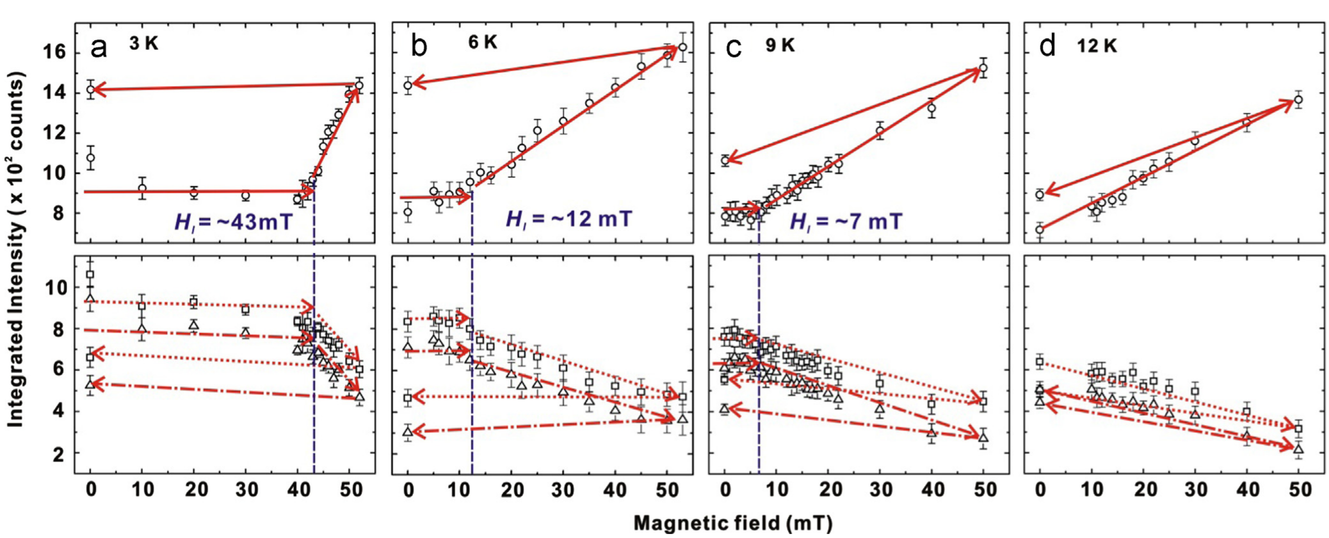

Fig. 4. Magnetic field dependence of the ferromagnetic peaks (1 0 1) (open circles), and satellite peaks, $(1\ 0\ 1)^-$; open squares and $(1\ 0\ 1)^+$; open triangles at (a) 3 K, (b) 6 K, (c) 9 K, and (d) 12 K. The arrows indicate the sequence of the magnetic fields applied to the sample and H_l represent the critical magnetic fields of the *lock-in* transition.

follows an exponential curve (solid line). At 12 K, where the satellites have diminished, the intensities of the Bragg peaks increase continuously with the magnetic field. Thus, the influence of an external field on the magnetic ordering should be described by the threshold (T≤11 K) or the non-threshold mode transition (T≥11 K) in the temperature dependence. From the field dependence of the Bragg peaks obtained at 9 K, the ICM–CM (FM) transition was exhibited at 200 mT, and from the temperature dependence of Bragg peaks measured without a magnetic field, the same transition (ICM–CM (FM)) was observed at 20 K.

The magnetic phase diagram shown in Fig. 5 represents the spin arrangement, and it may be applicable to the $Tb_{1-x}Er_xNi_5$ system for $x < 0.2$. The lock-in phase in the temperature domain (refer Fig. 1(c)) and lock-in phase in the magnetic field domain seem to be closely related with each other. Below the critical temperature (T_l) and the critical field (H_l), the system tends to keep its magnetic state until the temperature or the magnetic field exceeds the critical values, T_l and H_l, respectively.

This interesting magnetic behavior of the system, $Tb_{1-x}Er_xNi_5$ (x≤0.2), seems to be relevant with the competitive Tb–Tb exchange interactions between the first and next-nearest neighbors. This phenomenon suggests a possible application for devices with a weak magnetic field controlled switch or memory, although the operating temperatures in this system are quite low.

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