Elastic anomalies at successive phase transitions in NdRu$_2$Al$_{10}$

T Suzuki$^{1,2,3}$, I Ishii$^1$, Y Suetomi$^1$, H Muneshige$^1$, T K Fujita$^1$, S Tanimoto$^4$ and T Nishioka$^4$

$^1$Department of Quantum Matter, AdSM, Hiroshima University, Higashi-Hiroshima 739-8530, Japan
$^2$Institute for Advanced Materials Research, Hiroshima University, Higashi-Hiroshima 739-8530, Japan
$^3$Cryogenics and Instrumental Analysis Division, N-BARD, Hiroshima University, Higashi-Hiroshima 739-8526, Japan
$^4$Graduate School of Integrated Arts and Sciences, Kochi University, Kochi 780-8520, Japan

E-mail: tsuzuki@hiroshima-u.ac.jp

Abstract. We carried out ultrasonic measurements on NdRu$_2$Al$_{10}$ single crystals in order to clarify what kind of phase transition occurs in this compound. The elastic moduli were measured as a function of temperature from 0.5 K to 150 K using the phase comparison-type pulse echo method. As the temperature is decreased from 150 K, longitudinal elastic modulus $C_{11}$ and transverse elastic modulus $C_{44}$ show a monotonic enhancement. With further decreasing temperature, both elastic moduli show abrupt hardening at 2.4 K and then show two-step softening at 1.3 K and 1.0 K. We found obvious hysteresis around both transitions at 1.3 K and 1.0 K in the temperature sweep, whereas no hysteresis was observed around the transition at 2.4 K within our experimental accuracy. The abrupt hardening at 2.4 K is very similar to that found in CeRu$_2$Al$_{10}$ and CeOs$_2$Al$_{10}$, suggesting that the transition at 2.4 K originates from the same mechanism of the transitions in CeRu$_2$Al$_{10}$ and CeOs$_2$Al$_{10}$. This is the first report that NdRu$_2$Al$_{10}$ undergoes the successive phase transitions.

1. Introduction

The cage compounds with the composition formula $RT_2$Al$_{10}$ ($R$: rare earth; $T$ = Fe, Ru and Os), which have the YbFe$_2$Al$_{10}$-type orthorhombic structure, have attracted much attention because CeRu$_2$Al$_{10}$ and CeOs$_2$Al$_{10}$ undergo a novel phase transition at $T_0$ = 27.3 K and 28.7 K, respectively [1-3]. There have been many proposals about a mechanism of ordering. Nishioka et al. suggested a charge-density-wave (CDW)-like transition at $T_0$ in CeRu$_2$Al$_{10}$ [2]. Meanwhile, Tanida et al. proposed a magnetic-type phase transition in which the singlet spin pair between spins on neighboring Ce ions is formed below $T_0$ from the measurements on the thermal and transport properties of La-diluted Ce$_{1-x}$La$_x$Ru$_2$Al$_{10}$ [4]. Recently, an antiferromagnetic structure along the c-axis below $T_0$ is reported in inelastic neutron scattering and muon spin relaxation experiments [5].

We already measured temperature dependences of elastic moduli of CeRu$_2$Al$_{10}$ and reported that abrupt hardening occurs at $T_0$ in all elastic moduli without hysteresis, suggesting that the transition is of the second order and there is a $Q\varepsilon^2$-type coupling term in an effective Hamiltonian where $Q$ and $\varepsilon$ represent an order parameter and an elastic strain, respectively [6]. This elastic behaviour is almost identical with that of the typical CDW-transition-compounds TaS$_3$ and NbSe$_3$[7,8]. Therefore we...
pointed out that the plausible origin for the transition at $T_0$ in CeRu$_2$Al$_{10}$ is the CDW ordering [6].

From the optical conductivity measurements, Kimura et al. proposed that the phase transition in the same CeRu$_2$Al$_{10}$ originates from the CDW formation which induces antiferromagnetic ordering [9], supporting our conclusion for CeRu$_2$Al$_{10}$.

Very recently, successive phase transitions were found in NdOs$_2$Al$_{10}$ [10]. In order to investigate what kind of phase transition occurs in NdRu$_2$Al$_{10}$, we synthesized single crystals of NdRu$_2$Al$_{10}$ and measured elastic moduli using an ultrasonic technique. This is the first report on the successive phase transitions in NdRu$_2$Al$_{10}$.

2. Experimental

Single crystals of NdRu$_2$Al$_{10}$ were grown by the Al self-flux method. The lattice constants of NdRu$_2$Al$_{10}$ are referred from ref. 11, such as $a = 9.115$ Å, $b = 10.261$ Å and $c = 9.152$ Å. The elastic moduli $C_{11}$ and $C_{44}$ were measured as a function of temperature from 0.5 to 150 K by the phase comparison-type pulse echo method using a $^3$He cryostat with a superconducting magnet. The modulus $C_{11}$ is the longitudinal mode propagating along the $a$-axis, and $C_{44}$ is the transverse mode propagating along the $b$-axis with the polarization direction along the $c$-axis. The elastic modulus $C$ was calculated using $C = \rho v^2$ with a room-temperature mass density $\rho = 4.782$ g/cm$^3$, where $v$ is the sound velocity in a sample.

3. Results and discussion

Figure 1 shows temperature dependence of the longitudinal elastic modulus $C_{11}$ in NdRu$_2$Al$_{10}$, which was measured with the ultrasonic frequency of 99 MHz. The inset shows $C_{11}$ in an expanded scale below 3.5 K. The modulus $C_{11}$ is the linear response to the $\varepsilon_{xx}$ strain. The modulus $C_{11}$ increases monotonically with decreasing temperature at high temperatures. With further decreasing temperature, abrupt hardening occurs at $T_1 = 2.4$ K and then two-step softening appears around $T_2 = 1.3$ K and $T_3 = 1.0$ K, respectively.

![NdRu$_2$Al$_{10}$](image)

Figure 1. Temperature dependence of $C_{11}$

When we repeated the temperature sweep around three transitions, we found hysteresis around $T_2$ and $T_3$ in contrast to no hysteresis around $T_1$ in the temperature sweep. Here we report, for the first
time, that NdRu$_2$Al$_{10}$ undergoes three phase transitions: the second order transition at $T_1$ and the first order transitions at $T_2$ and $T_3$, respectively.

Figure 2 shows temperature dependence of the transverse elastic modulus $C_{44}$ in NdRu$_2$Al$_{10}$, which was measured with the ultrasonic frequency of 102 MHz. The modulus $C_{44}$ is the linear response to the $\epsilon_{yz}$ strain. This transverse elastic modulus shows almost the same temperature dependence of $C_{11}$, such as the monotonic increase at high temperatures, the abrupt hardening at $T_1$ and the two-step softening around $T_2$ and $T_3$. However, the hysteresis around $T_2$ and $T_3$ is more obvious than that in the $C_{11}$ mode. This may be related to the symmetry of ordering structure corresponding to the symmetry of the $\epsilon_{yz}$ strain.

![Figure 2. Temperature dependence of $C_{44}$](image)

At the second order transition at $T_1$, both of longitudinal and transverse moduli abruptly enhance without hysteresis, suggesting that there is a $Q\epsilon^2$-type coupling term in the effective Hamiltonian [12]. This elastic behaviour is almost identical with that in CeRu$_2$Al$_{10}$ [6]. Therefore, we estimate that this transition also originates from the CDW transition accompanied by antiferromagnetic ordering which was found in CeRu$_2$Al$_{10}$. The isomorphic compounds RT$_2$Al$_{10}$ may have similar instability commonly.

As for the first order transitions at $T_2$ and $T_3$, the origin is unclear at present. However, in our preliminary elastic measurements under magnetic fields, we found that both transition temperatures of $T_2$ and $T_3$ decrease with increasing the magnetic field. This may suggest that both transitions originate from an antiferromagnetic ordering. In order to clarify the origin of phase transitions, we need to perform experiments on magnetic properties of NdRu$_2$Al$_{10}$.

4. Conclusion
We carried out ultrasonic experiments on NdRu$_2$Al$_{10}$ single crystals to measure temperature dependence of longitudinal and transverse elastic moduli in the temperature range between 0.5 and 150 K. We found for the first time that NdRu$_2$Al$_{10}$ undergoes the second order transition at $T_1 = 2.4$ K and the first order transitions at $T_2 = 1.3$ K and $T_3 = 1.0$ K, respectively. We estimate that a CDW transition accompanied by antiferromagnetic ordering occurs at $T_1$. 

5. References

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Acknowledgments
This work was supported by a Grant-in-Aid (No. 20340093) and a Grant-in-Aid for Scientific Research on Innovative Areas "Heavy Electrons" (No. 20102005) from the Ministry of Education, Culture, Sports, Science and Technology of Japan.