Zero-field μSR measurements to investigate the magnetic ordering of $Nd_2Ru_2O_7$

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Pyrochlore oxides have a general formula $A_2B_2O_7$, where A and B represent trivalent rare-earth and tetravalent transition metal ions, respectively. The pyrochlore oxides are constructed from the interpenetrating corner-sharing tetrahedral lattices of A and B sites.[1)] The spins at vertices of the tetrahedral lattice are magnetically frustrated and can lead to various novel physical properties.[1)] The magnetic frustration, competition between the exchange and dipolar interactions, and crystal electric field effect control the nature of the ground state of pyrochlore oxide.[2)]

Pyrochlore ruthenate, $A_2Ru_2O_7$, has Ru $4d^4$ electrons in the low-spin state with $S = 1$. In $Nd_2Ru_2O_7$, both Nd and Ru are magnetic ions. The magnetic ground state of $Nd_2Ru_2O_7$ is an interesting research topic because we can investigate the coupling between Nd and Ru spins by comparing it with that of $Nd_2Ir_2O_7$ known to have Ir^{4+} ions with $5d$ electrons, and they show ferromagnetic coupling between Nd and Ir spins.[3)] $Nd_2Ru_2O_7$ showed magnetic anomalies around 1.8 K, 21 K, and 146 K.[4,5)] At 1.8 K and 146 K, the magnetic transition were attributed to the ordering of Nd and Ru spins, respectively.[4)] The origin of the anomaly at 21 K remains debatable.[4,5)] Accordingly, we investigated the magnetic properties of $Nd_2Ru_2O_7$ using various measurement techniques such as muon spin relaxation (μ^+SR) measurement.

The polycrystalline $Nd_2Ru_2O_7$ was prepared using a solid-state reaction method. μ^+SR experiments were carried out on an HiFi spectrometer at the ISIS, Rutherford Appleton Laboratory in the UK and on the ARTEMIS spectrometer at Material and Life Science Experiment Facility (MLF), J-PARC in Japan. We measured the μ^+SR time spectra in the zero-field (ZF) condition at a temperature range of 2–150 K on HiFi, whereas the time spectra below 5 K down to 0.3 K were obtained by using Heliox cryostat on ARTEMIS.

The oscillations do not occur in the ZF-μ^+SR time spectra of $Nd_2Ru_2O_7$, as shown in Fig. 1(a). However, the decreases in the initial asymmetry at $t = 0$ was observed by decreasing the temperature indicating the appearance of a magnetic ordered state. The time spectra were analyzed using two exponential functions as

$$A(t) = A_1 \exp(-\lambda_1 t) + A_2 \exp(-\lambda_2 t) \quad (1)$$

The first and second components of Eq. (1) correspond to the slow and fast components of muon spin relaxation, respectively, and λ represents the muon-spin relaxation rates.

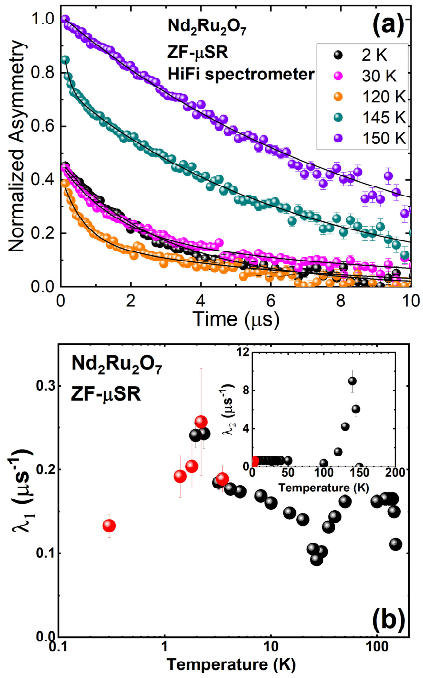

Fig. 1. (a) ZF-μ^+SR time spectra of $Nd_2Ru_2O_7$. (b) Temperature dependence of λ_1; the inset shows the temperature dependence of λ_2.

The appearance of Ru ordering at 146 K is confirmed from a sharp peak observed in $\lambda_2(T)$, as shown in the inset of Fig. 1(b). λ_1 decreases below 50 K with decreasing temperature and shows a dip around 30 K; this anomaly is not related to the anomaly at 21 K observed from the DC susceptibility measurement, which was expected from the magnetic property of Nd_3RuO_7.[6)] Therefore, the anomaly around 30 K in the temperature dependence of λ_1 could be related to the short-range magnetic interaction of Nd spins. The long-range ordering of Nd spins was expected to appear around 2 K, as indicated by a peak in the temperature dependence of λ_1. Currently, we cannot determine the magnetic interaction between Nd and Ru spins. Further investigation using continuous muon beam is required to determine the internal field as a function of temperature that occurred in this system.

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