Spin dynamics in Pyrochlore $Nd_2Mo_2O_7$

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$Nd_2Mo_2O_7$ is metallic and exhibits a ferromagnetic transition at a Curie temperature $T_c = 93$ K. In contrast with other Nd-pyrochlore systems, such as $Nd_2Sn_2O_7$ and $Nd_2Zr_2O_7$, where the magnetic moments are localized, $Nd_2Mo_2O_7$ possesses delocalized electrons, which may host novel phenomena associated with the interaction between the localized moments Nd^{3+} and the itinerant moment Mo^{4+}. Particularly, $Nd_2Mo_2O_7$ exhibits unusual spin chirality,[1,2] and possesses a two-in-two-out (AIAO) spin ice spin configuration on Nd^{3+}. Unlike the AIAO magnetic order of Ir^{4+} in $Nd_2Ir_2O_7$, the XY type order of Ru^{4+} in $Nd_2Ru_2O_7$, and Mo^{4+} in $Nd_2Mo_2O_7$ orders along the 001 direction. In $Nd_2Mo_2O_7$, the anomalies appearing at 93 K in both the magnetic and heat capacity is attributed to the Mo^{4+} ordering temperature, whereas the ordering of Nd^{3+} becomes significant below 30 K.[3] Therefore, it is interesting to study the spin dynamics in the delocalized electron system of $Nd_2Mo_2O_7$ and compare them to those of other Nd-pyrochlores by μSR technique. Moreover, applied fields in $Nd_2Mo_2O_7$ may change the ground state of Nd^{3+}.[3] μSR experiments were carried out at ARGUS spectroscopy, RIKEN-RAL, between the temperature ranges of 5–300 K to obtain the information below and above the magnetic transition temperatures of 30 and 93 K. Figure 1(a) shows the zero-field mSR relaxation spectra at 7.5, 35, 95, and 160 K and the fitted curves using the function of $A_1 \exp(-\lambda_1 t) + A_2 \exp(-\lambda_2 t) + A_3$. The plots of the fitted parameters are shown in Fig. 1(b). The initial drop of the muon relaxation below 95 K indicates the magnetic order of the compound, which can also be confirmed by the increasing of the relaxation rate λ_1 in Fig. 1(b).

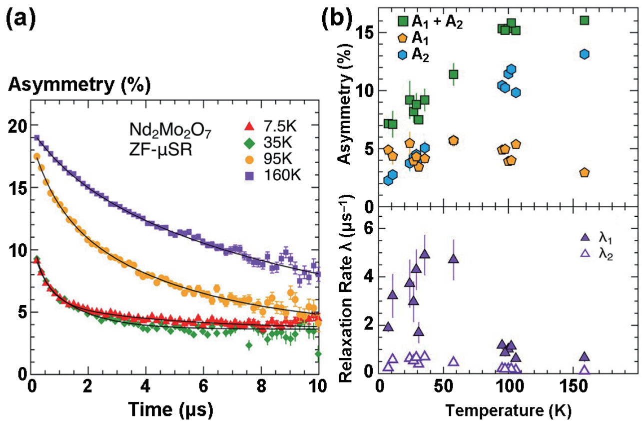

Fig. 1. (a) zero-field μSR relaxation spectra at 7.5, 35, 95, and 160 K and fitted curves using the function of $A_1 \exp(-\lambda_1 t) + A_2 \exp(-\lambda_2 t) + A_3$; (b) plots of the fitted parameters.

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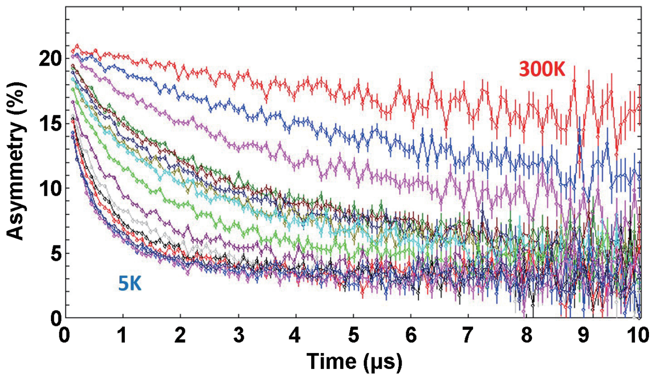

Fig. 2. Temperature-dependence of the asymmetry spectra of muon relaxation signal between 5 and 300 K in the applied field of 4000 G. The spectra can be separated into three different temperature intervals of 30 K and 93 K, the transition temperatures of Nd^{3+} and Mo^{4+} ordering temperatures, respectively.

The initial drop made it impossible to determine the magnetic volume fraction of the AIAO spin structure. However, the transition at 30 K cannot be picked up clearly by muon relaxation. The asymmetry of the slow relaxation parameter exhibits a magnetic anomaly. Longitudinal fields were applied up to 4000 G to evaluate the decoupling of muons on the sites. An applied constant of 4000 G in the temperature-dependent scans was also utilized in the experiments. The transitions at 30 and 93 K can be clarified in the measurements as shown in Fig. 2, in which the relaxations of the spectra are separated to three groups in three different temperature ranges. Proper models have to be proposed to fit these curves and higher fields are necessary to decouple the muon relaxations for further study on this compound.

At 5 K, the significant differences of the asymmetry and muon relaxation have been observed before and after an applied field (not shown in the figure). Similar behaviour does not appear at a higher temperature of 85 K. This hysteresis at 5 K indicates ferromagnetic components at low temperature. In our experiments, this hysteresis even happens in a small applied field of 100 G. The magnetic structure of $Nd_2Mo_2O_7$, had been reported as a spin chirality "umbrella" structure,[4] which is consistent with the observation made in the μSR spectra at low temperatures. This phase occurs below the Nd^{3+} ordering temperature only, and does not exist below the Mo^{4+} ordering temperature between 93 K and 30 K. However, muon precession was not observed in the experiments. We are working on a proper model to fit the spectra below 30 K.

References
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