Magnetic properties of Alkali-metal Superoxide, NaO_2

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Magnetism in the p-electron system has attracted attention for the possibility of new types of magnetic informative materials. Alkali-metal superoxides, AO_2 (A = Na, K, Rb, Cs), present an interesting example of magnetic materials on the basis of unpaired p-electrons. The magnetic ordering of KO_2, RbO_2, and CsO_2 have been observed at temperatures 7 K, 15 K, and 9.6 K, respectively using specific heat.[1] However, the magnetic ground state in NaO_2 is not yet clarified.

Clear anomalies from the measurement of magnetic susceptibility were observed at a temperature between 230 and 200 K in NaO_2. These anomalies were consistent with the expected structural phase transition, as discussed in the previous study.[1] The magnetic susceptibility, χ, drops sharply below 40 K, as shown in Fig. 2 (a). We checked that the sample quality was good and the anomalies in magnetic susceptibility could be reproduced in all NaO_2 sample batches (see sample condition in Fig. 1). The sudden change of susceptibility value toward zero is an indication of spin gap state as observed in other spin gap systems, $NaTiSi_2O_6$ and TiOCl.[2,3] Mahanti *et al.* reported that NaO_2 has some similarities with one-dimensional spin system.[4]

There was no emergence of muon-spin precession from the previous zero-field (ZF) μSR experiment in NaO_2 down to 0.3 K measured at the RIKEN-RAL Muon Facility and DOLLY PSI, which indicated the absence of magnetic ordering. To further study the magnetic properties in NaO_2, we measured the depolarization rate in ZF close to anomaly $\sim$ 40 K (observed by magnetic susceptibility) using CHRONUS spectrometer at the RIKEN-RAL Muon Facility.

The temperature dependence of exponential relaxation rate, λ, measured using ZF- μSR showed an anomaly at a temperature below 40 K that was significantly consistent with the anomaly observed by magnetic susceptibility measurement, as shown in Fig. 2 (a). This indicated the possibility of magnetic transition around this temperature.[5] The red line in Fig. 2 (b) is the fitting line that utilizes the following function: $\lambda(\mathrm{T}) = \lambda_0/[1 + A\exp(\frac{-2\varDelta}{T})]$, in which A is the constant and $\varDelta$ is the spin-gap value.[6] The obtained result of $\varDelta$ is estimated to be $\sim$ 35 K. The possibility of the formation of spin gap in NaO_2 must be further explored using other experimental techniques.

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Fig. 1. Some sample batches of NaO_2 used for μSR experiment. Labels indicate the number of sample batches. Bright and dense yellow color imply that sample quality is good.

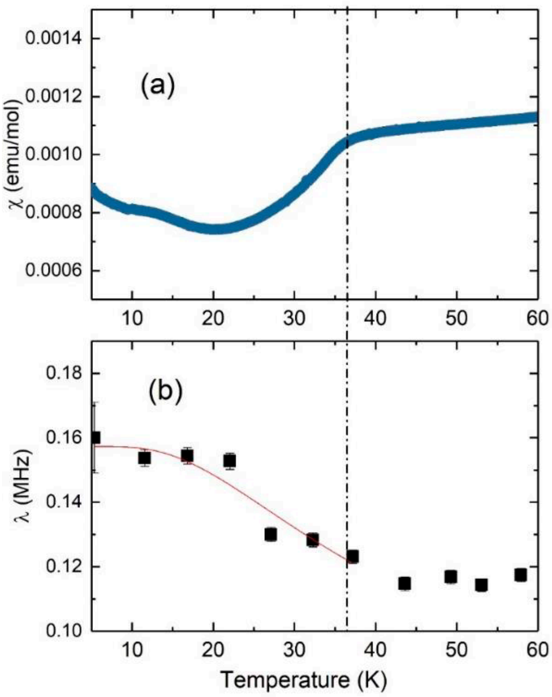

Fig. 2. (a) Magnetic susceptibility of NaO_2 and (b) Temperature dependence of exponential relaxation rate, λ, measured using ZF-μSR at various temperature above and below 40 K.

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