Study of Magnetic Ordering by p-orbital in RbO_2 using μSR

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Alkali metal superoxides AO_2 (A= Na, K, Rb, Cs) present an interesting example of magnetic materials based on p-elements. This become the first example of an inorganic quantum spin system with unpaired π-electrons.[1] Alkalimetal superoxides adopt the rocksalt-type crystal structure and two oxygen atoms form a dumbbell shaped structure sharing one excess electron, $O_2{}^-$, which is known as the "superoxide" anion. This leads to one unpaired spin (S=1/2) in a pair of degenerate π^* (antibonding) molecular orbitals. The magnetic ordering of KO_2, RbO_2, and CsO_2 have been observed at temperatures of 7 K, 15 K, and 9.6 K, respectively by using specific heat measurement.[2] The Tomonaga Luttinger Liquid (TLL) model suggested for CsO_2 is supposed to present a field-induced magnetic order related to the TLL state.[3]

Therefore, detailed investigation on the magnetic properties near or in the zero-field (ZF) condition is strongly required to describe the magnetically ordered state appearing in CsO_2 and other alkali metal superoxides. Last year, we have carried out μSR measurements in RbO_2 at the RIKEN-RAL muon facility by using the pulsed muon beam. At that time, we felt that the sample quality was not so good. For that reason, in the next beam time, we improved the sample quality and measured the new batch of RbO_2 samples.

No clear muon-spin precession was seen at any temperature (Fig. 1), however the decrease in the initial asymmetry around the suggested T_N was clearly observed. The anomaly was also observed at ~15 K as shown in Fig. 2(a).

The asymmetry parameter can represent the magnetic volume fraction. By comparing the asymmetry parameters in Fig. 2(b), it seems that the new sample has bigger magnetic volume fraction than the old sample. It means that we succesfully improved the sample quality.

The decrease in the initial asymmetry possibly means that the magnetically ordered state appears causing the depolarization behavior faster than the observable limit of the pulsed muon facility. This ordered state might accomodate the fast muon-spin precession. Therefore, it is indispensable to test RbO_2 by using the continuous muon beam in order to detect clear evidence of the appearance of magnetically ordered states. As we expected, the result of μSR measurement at PSI, Switzerland, by using continuous muon beam showed clear-muon spin precession at the temperature ~15 K, indicating long-range magnetic ordered state (the result is not shown in this report).

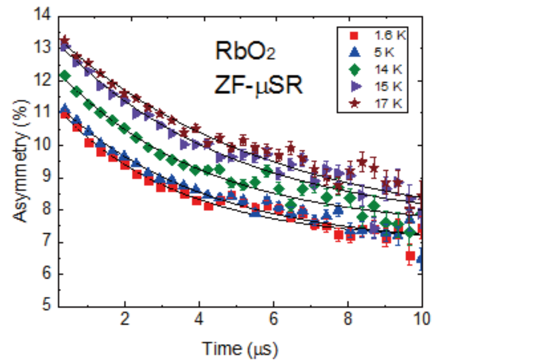

Fig. 1. ZF-μSR time spectrum for RbO_2 for the first 10 microsecond from 17 K down to base temperature.

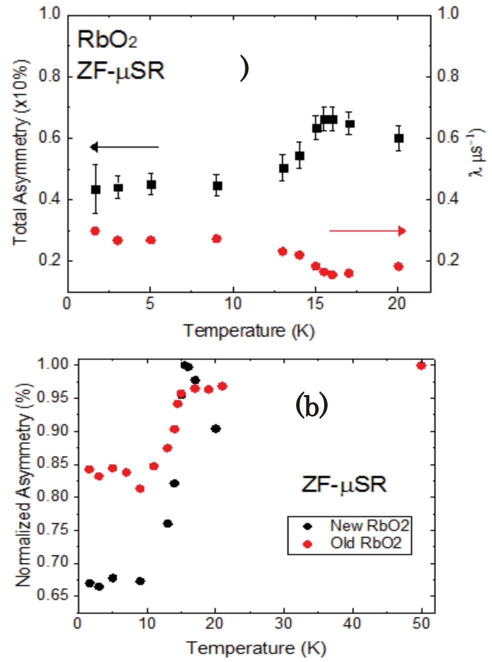

Fig. 2. (a) Temperature dependence of the initial asymmetry and relaxation rate of the ZF-μSR time spectrum measured at the RIKEN-RAL Muon Facility. The anomaly in the μSR measurement is observed at 15 K around the suggested T_N. (b) Comparison between the new and old RbO_2 samples. The result of the old sample has been reported in RIKEN APR 2016 Vol.50.

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