

Magnetic ordering and spin dynamics driven by p -orbital in RbO_2

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Magnetism in the π -electron system has attracted attention for the possibility of the new kinds of the magnetic informative materials. Alkali-metal superoxides AO_2 ($A = \text{Na}, \text{K}, \text{Rb}, \text{Cs}$) present an interesting example of magnetic materials on the basis of p -elements. These systems have a dumbbell-type bonding state of O atoms forming the valence state of O_2^- and resulting in one unpaired π -electron on the O_2^- dumbbell. They further show the changing of crystal structure introducing the splitting of the p -orbital degeneracy, similar to the Jahn-Teller effect. Beside the crystallographic phase transition due to molecular ordering of the disordered O_2^- , the magnetic order in alkali metal superoxide is interesting to study. In the case of superoxides, the number of unpaired electrons is only one on the O_2^- dumbbell, and magnetic superexchange interaction is expected between those unpaired spins through the A metal. Accordingly, a different magnetically ordered state from that observed in the solid oxygen molecule¹⁾ is expected in superoxides, but detailed information on magnetic properties is still missing. 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, using specific heat measurement.²⁾ The Tomonaga Luttinger Liquid (TLL) model suggests that a field-induced magnetic order should appear in the CsO_2 that is related to the TLL state.³⁾

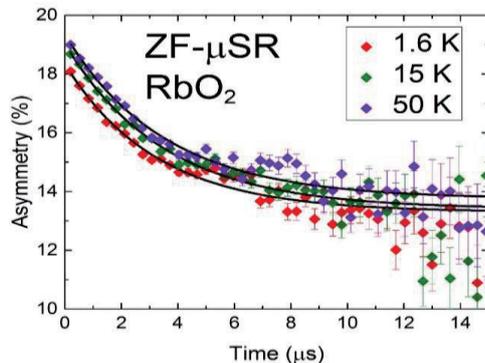


Fig. 1. ZF- μ SR time spectra for CsO_2 for the first microsecond from 10 K down to base temperature.

Therefore, a detailed investigation on the magnetic properties near or in the zero-field (ZF) condition is strongly required to describe the magnetically ordered state that appears in the CsO_2 and other alkali metal superoxides.

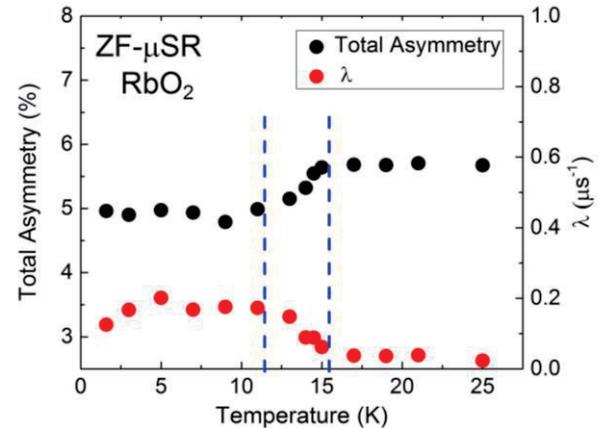


Fig. 2. Temperature dependence of the initial asymmetry and relaxation rate (λ) of the ZF- μ SR time spectra measured at the RIKEN-RAL Muon Facility. The anomaly in the μ SR measurement is observed in between 10 and 15 K around the suggested T_N .

We carried out μ SR measurements in CsO_2 at the PSI Switzerland using the continuous muon beam. Clear spontaneous muon-spin precession behavior indicates the appearance of a long-range magnetic ordered state. This is evidence of the coherent static magnetically ordered state of π -electrons in oxygen molecules. Another alkali-metal superoxide, RbO_2 , which has a crystal structure similar to that of CsO_2 (CaC_2 -like), was tested at the RIKEN-RAL. In this system, only a type of anomaly in the magnetic susceptibility of RbO_2 is reported at $T_N \sim 15$ K, as indicated.⁴⁾ Unfortunately, we could not observe clear muon-spin precession as shown in Fig.1, although the decrease in the initial asymmetry around the suggested T_N was observed, as displayed in Fig. 2. The decrease in the initial asymmetry and increase in the relaxation rate (λ) possibly means the magnetically ordered state appears, causing a depolarization behavior that is faster than the time limitation of the pulsed muon facility. This ordered state might accommodate the fast muon-spin precession as well as the case of CsO_2 . Therefore, it is necessary to test RbO_2 at PSI using the continuous muon beam in order to detect clear evidence of the appearance of magnetically ordered states.

References

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