μSR study on ferrimagnetism of Na-K alloy clusters incorporated into zeolite LSX under high-pressure helium gas

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Various types of magnetic orderings such as ferromagnetism, antiferromagnetism, and ferrimagnetism have been found in three dimensionally arrayed alkali-metal nanoclusters in zeolite crystals.[1)] The magnetic properties depend on the structure of the zeolite, species of the alkali element, and electron density of the clusters. They are a new class of magnetic materials because their magnetic orderings are realized by the mutual interactions between the s-electrons and they contain no magnetic elements.

In zeolite LSX (low-silica X), β-cages with an inner diameter of 7 Å are arrayed in a diamond structure. A supercage with an inner diameter of 13 Å is formed between the β-cages. The supercages are also arrayed in a diamond structure. It is known that an N-type ferrimagnetism appears when Na-K alloy clusters are incorporated into zeolite LSX ($Na_4K_8Al_{12}Si_{12}O_{48}$ per supercage) by loading K atoms for a certain range of the loading density. The highest Curie temperature T_C is approximately 20 K. Recently, we found that a new ferromagnetic phase with T_C as high as 60 K is formed on applying pressure using helium gas. Because the formation of the new magnetic phase has a slow time dependence and is irreversible, it is speculated that the helium atoms are loaded into the pores of the zeolite crystal by pressure and directly change the electronic states of the clusters resulting in the enhancement of the magnetism. In this work, we performed μSR measurements on this system under a high-pressure helium gas to investigate the new magnetic phase.

A high pressure cell newly made of Ti alloy was loaded with a powder sample of K-loaded LSX. This sample shows a ferrimagnetic ordering below 20 K at ambient pressure. μSR measurements were performed at the RIKEN-RAL Muon Facility in the U. K. The pressure cell was loaded into the VARIOX cryostat and connected to a high-pressure He gas handling system which allowed us to control the pressure at any temperatures.[2)] We used a double-pulsed decay muon beam with a momentum of 64.3 MeV/c. Zero-field μSR spectra were obtained by utilizing the ARGUS spectrometer.

The obtained spectra showed slow relaxations. At 50 MPa, the initial asymmetry A_0 of the spectrum gradually decreased below $\simeq$ 90 K and quickly below $\simeq$ 20 K, as shown in Fig. 1. The decrease in A_0 indicates that the muon spin is depolarized within the time resolution of the measurement, $\simeq$ 0.4 μs, owing to the appearance of a strong internal magnetic field. After reducing the pressure, the decrease in A_0 below $\simeq$ 90 K disappeared, as seen in Fig. 1. Therefore, the gradual decrease in A_0 at 50 MPa can be attributed to the high T_C magnetic phase. The phase transition is not sharp. This may be due to a distribution of T_C originating from a certain inhomogeneity of the He atom loading. At 50 MPa, the ambient pressure phase with $T_C \simeq$ 20 K still remained. thereby indicating a phase separation. A part of the crystal may be loaded with He atoms by pressure. The amplitude of the decrease in A_0 for the high T_C magnetic phase is approximetely half of that in the 20-K phase at an ambient pressure. This result indicates that although there are some inhomogeneities, the high T_C magnetic phase has sufficient volume and can be regarded as bulk magnetism, which does not originate owing to any impurities.

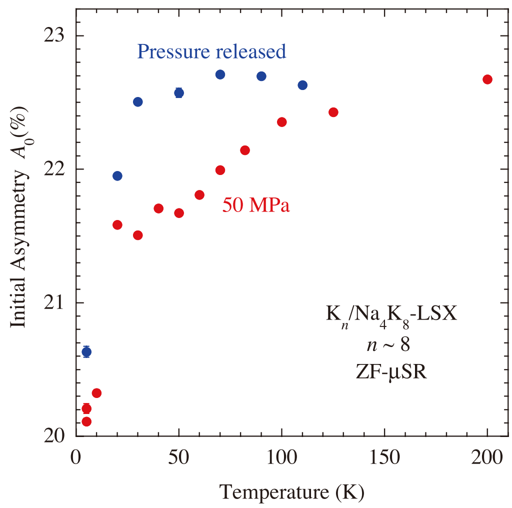

Fig. 1. Initial asymmetry of the μSR spectra of Na-K alloy clusters in zeolite LSX as a function of temperature. The red circles are data taken at 50 MPa and the blue circles are those after reducing the pressure.

References
1) T. Nakano, Y. Nozue, Adv. Phys. X **2**, 254 (2017).
2) I. Watanabe, Y. Ishii, T. Kawamata, T. Suzuki, F. L. Pratt, R. Done, M. Chowdhury, C. Goodway, J. Dreyer, C. Smith, M. Southern, Physica B **404**, 993 (2009).

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