7-1 First Observation of ^{239}Pu Nuclear Magnetic Resonance

− Elucidation of the Electronic State in Pu Compounds −

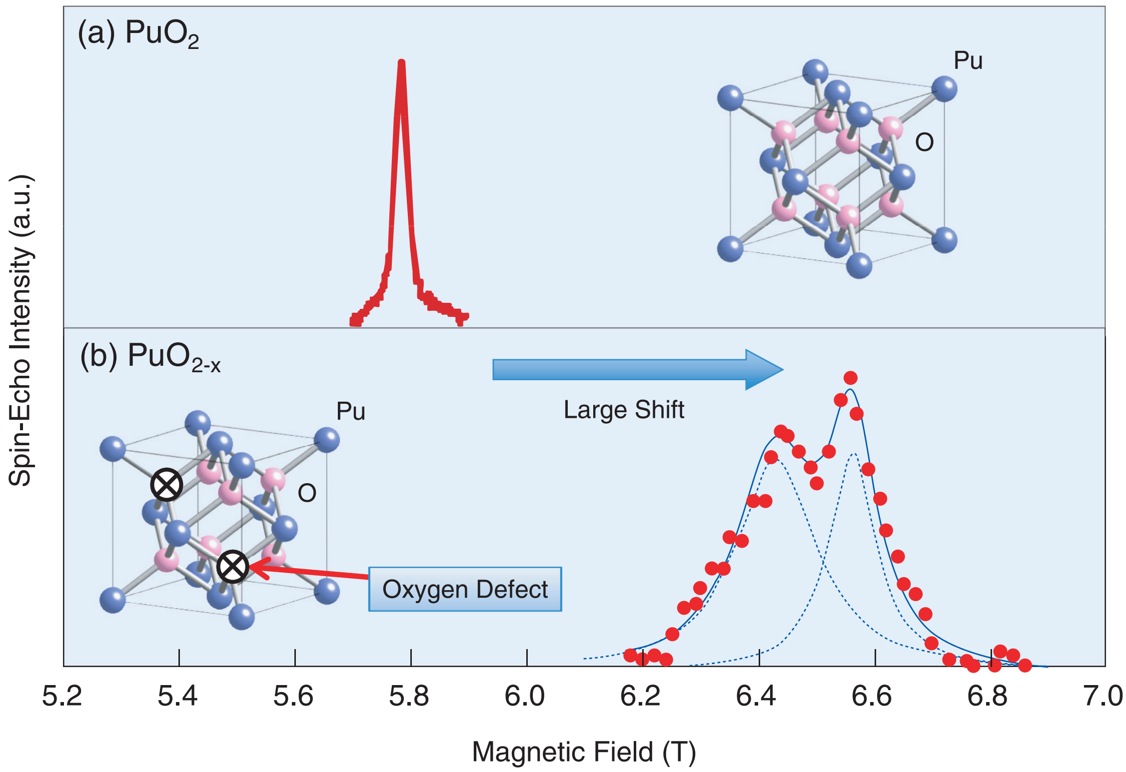

Fig.7-3 ^{239}Pu NMR signals observed for the first time in the world
^{239}Pu NMR spectra in (a) pure PuO_2 and (b) PuO_{2-x}, which has oxygen defects. Insets of (a) and (b) show the crystal structures of PuO_2. The solid and dotted blue curves represent the deconvoluted Gaussian curve fit to the data and each component, respectively.

Nuclear magnetic resonance (NMR) can be used to explore electronic states via the interaction between nuclear and electron spins. NMR has been applied to a wide range of fields, such as molecular structure analysis in chemistry and biology, for materials science, and for magnetic resonance imaging (MRI) in medicine. Since an NMR signal was first observed in 1946, it has become possible to detect more than 90 types of nuclear isotopes. Nevertheless, NMR analysis of the actinide nuclei has not been possible, except for ^{235}U. Among the actinide elements, Pu has attracted much attention owing to its noble and mysterious character. Moreover, Pu is important as an atomic-energy fuel, but the electronic state of Pu ions has not been well understood. For such reasons, scientists around the world have been searching for the Pu NMR signal over the past 50 years or more, but until recently, it remained elusive.

We therefore initiated a ^{239}Pu NMR signal search through an international collaboration with the U.S. Los Alamos National Laboratory (LANL). After establishing a detailed experimental design, we succeeded in observing the ^{239}Pu NMR signal for the first time. This success stems from the precise control of many experimental parameters and the use of a very pure sample synthesized by chemists at LANL.

Fig.7-3(a) shows the ^{239}Pu NMR spectrum measured for high purity PuO_2. From the field dependence of the central NMR frequency, the nuclear magnetic moment, which is the important physical constant, has been implied to be 0.15 μN (μN is the nuclear magneton). Fig.7-3(b) shows the ^{239}Pu NMR spectrum measured in PuO_{2-x}, which has oxygen defects. When the two spectra are compared, it can be seen that the resonance frequencies and the structures are different. This result implies that the ^{239}Pu NMR analysis has high resolution and is very sensitive to the differences in the coordination state of oxygen.

With this success, it is expected that the research regarding the electronic state of various Pu compounds using ^{239}Pu NMR analysis will now progress. In particular, because the stability of the Pu ionic state in oxides can be judged by elucidating the electronic state of Pu, it is expected that ^{239}Pu NMR will be useful for constructing a safer, prolonged storage method for used atomic-energy fuel containing Pu, which is a globally important issue today.

Reference

Yasuoka, H., Chudo, H. et al., Observation of ^{239}Pu Nuclear Magnetic Resonance, Science, vol.336, no.6083, 2012, p.901-904.