6-4 Probing Interstitial Hydrogen States Using Positive Muons

— For Detailed Understanding of Rare-Earth-Based Hydrogen-Absorbing Alloys —

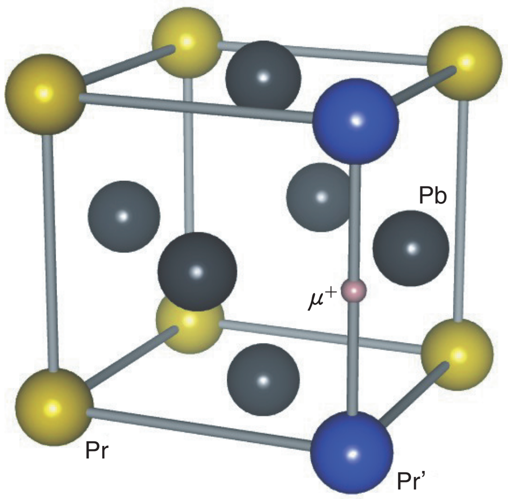

Fig.6-9 Crystal structure of $PrPb_3$ and μ^+ site
The implanted μ^+ stops at the midpoint between two Pr ions (Pr') and influences the f-electronic state of those ions.

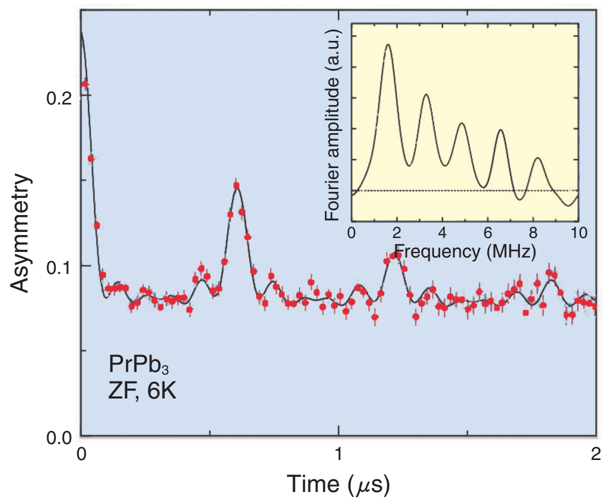

Fig.6-10 μ^+SR spectrum of $PrPb_3$ at 6K in zero magnetic field
The horizontal and longitudinal axes indicate time from arrival of the μ^+ and asymmetry of the positron emission relative to the μ^+ spin polarization, respectively. The inset shows a result of Fourier transform.

In recent years, much interest has been focused on hydrogen-based energy systems. The key of this technology is high-density storage of hydrogen, and hydrogen-absorbing (HA) alloy is useful for this purpose. The HA alloy absorbs the hydrogen to form metal hydride. In this process, the hydrogen molecule is dissociated into H atoms and these are stabilized at an interstitial site of the lattice. Detailed understanding of the interstitial H state is thus important for further improvement of HA properties.

We studied the interstitial H state in a material related to the HA alloy using positive muons (μ^+). The μ^+ can be regarded as a light isotope of ^{1}H since it possesses charge $+e$ and mass ~$1/9m_p$, where m_p is the mass of a proton. Therefore, chemical properties of the μ^+ in condensed matter are considered to be identical to those of ^{1}H except for isotope effects. The μ^+ implanted into materials stops at the interstitial site and decays into a positron and two neutrinos. The positron is preferentially emitted in the direction of the μ^+ spin, which evolves via magnetic interactions with surrounding electron and nuclear spins. Detailed analysis of the time evolution of the μ^+ spin provides us with information on the magnetic environment at the μ^+ site.

We launched this research project with investigation of the interstitial H state in a rare-earth-based intermetallic compound $PrPb_3$, related to the typical HA alloy $MmNi_5$ (Mm: mixture of light rare-earth elements). First, the μ^+ site in $PrPb_3$ was determined to be the midpoint between two Pr ions (Pr') as shown in Fig.6-9, using the μ^+ spin rotation and relaxation method (μ^+SR) in a high magnetic field. μ^+SR measurements in zero magnetic field were also performed, and the characteristic spectrum shown in Fig.6-10 was obtained in the paramagnetic state. This spectrum indicates quantization of the magnitude of a hyperfine field that is generated at each μ^+ site. From detailed analysis, it was clarified that coupling between Pr' and μ^+ spins is anisotropically enhanced as a result of deformation of the *f*-electron state owing to the μ^+ charge. The quantum character of the hyperfine field strongly supports this conclusion.

The present result suggests the importance of the electrostatic interaction between the *f*-electrons and the interstitial H in HA alloys. We plan further μ^+SR studies in HA alloys which have commercial promise, to clarify the relation between this interaction and the HA properties.

Reference

Ito, T. U. et al., Quantized Hyperfine Field at an Implanted μ^+ Site in $PrPb_3$: Interplay between Localized *f* Electrons and an Interstitial Charged Particle, Physical Review Letters, vol.102, issue 9, 2009, p.096403-1-096403-4.