Observation of Unusual Behavior in ^{55}Mn NQR for MnII site in β-Mn Metal

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Abstract

β-Mn metal is known not to show any magnetic ordering until low temperature, which is ascribed to an itinerant nearly antiferromagnet or a spin liquid state due to the geometrical frustration at MnII site. We have observed the sudden disappearance of the MnII-NQR signal accompanying by the rapid increase of the nuclear spin-lattice relaxation rate, T_1^{-1} above about 200 K. The behavior $T_1^{-1} \propto \sqrt{T}$ below about 100 K is consistent with the self-consistent renormalization theory for itinerant nearly antiferromagnet, however, the unusual behavior described above is difficult to explain in this framework.

Key words: β-Mn metal; NQR; nearly antiferromagnet; frustration

The high-temperature β phase of Mn metal, β-Mn, is one of a few examples of itinerant antiferromagnet. β-Mn has two crystallographically inequivalent sites. Eight atoms occupy site I and twelve atoms occupy site II in the twenty atoms per unit cell. The electronic state of Mn atoms at site II is expected to be more magnetic than at site I since the atoms at site II are more isolated than the atoms at site I.

For a long time, β-Mn has been classified as an itinerant nearly antiferromagnet, because it does not order magnetically until low temperature [1]. By the substitution of various impurities, the magnetic property changes from nearly antiferromagnet to weakly antiferromagnet, and further to antiferromagnetic metal because the magnetic property is strongly affected by the change of lattice constant and the number of d electrons [2]. Then β-Mn alloy has been considered to be a good model system to investigate the general property of itinerant antiferromagnet. However, it is proposed recently that no magnetic ordering of pure β-Mn metal is ascribed to a spin liquid state due to the geometrical frustration at MnII sites with three-dimensional network of corner-sharing regular triangles [3].

NMR measurements had been confined to MnI site until recent discovery of NQR signal from MnII sites [4]. We have carried out NQR measurements on ^{55}MnII in order to investigate the magnetic properties of β-Mn. Measurement at MnII site is more straightforward to study the magnetic properties in β-Mn metal.

β-Mn were obtained by quenching the ingots into ice water after annealing at 900 °C for 12 h. The ingots were crushed into powder with the particle size of less than 53 μm. NQR measurements were performed using a conventional spin echo technique.

Figure 1 shows the ^{55}Mn NQR spectra at 1.5 - 300 K for MnII site. Two lines around 4.5 and 9 MHz come from the transitions for $\pm 1/2 \leftrightarrow \pm 3/2$ and $\pm 3/2 \leftrightarrow \pm 5/2$, respectively. We found the two unusual behaviors as follows; (a) below about 100 K, the fine structure with small peak splitting grows up, and (b) NQR signal disappears completely above about 250 K.

At first, we mention to the unusual behavior (a). From the shape analysis of the upper and lower NQR lines, some small portion of MnII sites has a small in-

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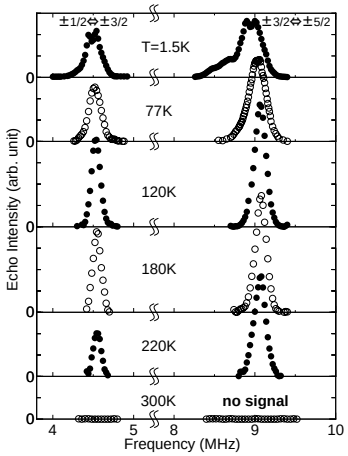

Fig. 1. Temperature dependence of ^{55}Mn NQR spectrum arising from Mn atoms at site II.

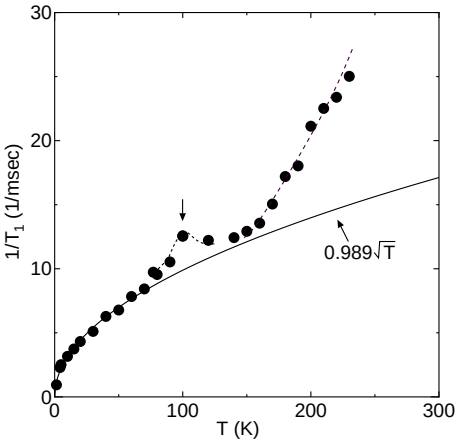

Fig. 2. Temperature dependence of T^{-1} of ^{55}MnII site.

ternal field of about 59 G at 1.5 K. The internal field decreases with increasing temperature and disappears at about 100 K, which suggests the magnetic order occurring at 100 K in some part in the sample. The small enhancement of the nuclear spin-lattice relaxation rate T_1^{-1} corresponding to the critical slowing down also appears as shown in Fig. 2 by the arrow. As the ordering temperature 100 K is close to the Neel temperature T_N=95 K of α-Mn, one may consider the order as a contamination effect of α phase. We intentionally prepared several samples possessing different quantity of α phase. The NQR spectra did not show any systematic change in theses samples, hence the possibility of the contamination effect of α-phase is ruled out. Alternatively, as the fine structure fades out a little in a larger powder particle size of 300 - 500 μm, the fine structure may have a parasitic origin from some region near the sample surface.

Next we mention to the unusual behavior (b). The disappearance occurs without any change of the quadrupole parameters of electric field gradient at MnII site, the quadrupole frequency ν_Q ($\simeq$ 4.46 MHz) and the asymmetry parameter η ($\simeq$ 0.01). Thus the unusual disappearance is not owing to any structural transition, but pure dynamical effect accompanying by the rapid increase of T_1^{-1} and hence the rapid increase of the spin echo decay rate T_2^{-1}. Figure 2 shows the temperature dependence of T_1^{-1}. Here we estimate T_1 assuming the magnetic relaxation since the magnetization recovery for upper and lower NQR lines show good fit to the recovery functions expected [5].

As shown in Fig. 2, T_1^{-1} follows $0.989\sqrt{T}$ (msec)$^{-1}$ between 1.5 K and 80 K, being consist with the previous result [4]. The $\sqrt{T}$ dependence of T_1^{-1} is consistent with the self-consistent renormalization (SCR) theory for spin fluctuations in itinerant nearly antiferromagnet [6]. However, T_1^{-1} significantly deviates upward from the $\sqrt{T}$ line above about 150 K after the small hump around 100 K. The unusual behavior of the rapid increase of T_1^{-1} seems to be difficult to explain within the framework of SCR theory.

Although we have no definite explanations for the unusual behavior at present, some causes may be considered. Volume expansion at high temperature may enhance the low frequency spin fluctuations because β-Mn is close to the critical boundary between nearly and weakly itinerant antiferromagnet. Additional relaxation through phonon coupling may contribute at high temperature. The break down of the spin liquid state above 150 K may be an origin of the unusual increase of T_1^{-1}. Impurity substitution and pressure effects are studying now to clarify the unusual behavior.

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