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Polarized Neutron Scattering Study of β-Mn and β-$Mn_{0.9}Al_{0.1}$

Masayuki SHIGA, Hiroyuki NAKAMURA, Masakazu NISHI[1]
and Kazuhisa KAKURAI[1]

Department of Metal Science and Technology,
Kyoto University, Kyoto 606-01
[1]*The Institute for Solid State Physics,*
The University of Tokyo, Tokyo 106

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Polarized neutron scattering measurements have been performed on β-Mn and β-$Mn_{0.9}Al_{0.1}$ to study spin fluctuations. A large magnetic scattering peak at around $Q=1.5$ Å^{-1} was observed in the spectra obtained in the quasi-elastic condition, indicating the existence of strong spin fluctuations with antiferromagnetic correlation. The energy and Q spectra at 290 K exhibit nearly the same profiles and intensity for both samples. However, the energy spectra at low temperatures are quite different. For the pure β-Mn, the intensity decreases with decreasing temperature and the energy width remains as broad as at 290 K. For β-$Mn_{0.9}Al_{0.1}$, on the other hand, the intensity increases due to narrowing of the energy spectrum at low temperatures, implying a slowing down of spin fluctuations. These results are discussed in terms of a spin-liquid to spin-glass transition caused by Al substitution.

[manganese, aluminum, polarized neutron scattering, spin fluctuations, frustration, spin liquid, spin glass]

β-Mn has a nearly temperature-independent susceptibility and does not show any magnetic ordering down to the lowest temperature, indicating the absence of local magnetic moment. However, there is much evidence which indicates the existence of strong spin fluctuations: a large electronic specific heat coefficient, γ,[1] and characteristic temperature dependence of the nuclear spin relaxation time suggesting antiferromagnetic spin fluctuations.[2,3] Recently, we investigated the effects of Al substitution on the magnetism of β-Mn and showed that substitution of a small amount of Al for Mn results in formation of local moments on Mn atoms and a spin-glass state at low temperatures.[4] Such a drastic change of magnetism was also observed in the $Y(Sc)(Mn_{1-x}Al_x)_2$ system.[5] Noting a special crystal structure of C15, we explained this behavior in terms of frustration of magnetic interactions.[5] It was shown that the ground state of $Y(Sc)Mn_2$ may be regarded as a quantum spin-liquid realized through the frustration, and that the substitution of Al for Mn causes a spin-liquid to spin-glass transition as a result of partial removal of spin configurational degeneracy. In other words, the spin fluctuations in this system transfer from very dynamical (quantum) fluctuations to more static ones maintaining antiferromagnetic correlations. It is possible that the Al substitution for β-Mn gives the same effect on the magnetism, although the crystal structure of β-Mn is more complicated and it may not be an ideal frustrated system. In order to clarify the character of spin fluctuations in this system, we have carried out inelastic neutron scattering measurements on β-Mn and β-$Mn_{0.9}Al_{0.1}$ by using polarized neutrons. In this letter, we report the results of measurements and discuss the origin of giant spin fluctuations in β-Mn.

A sample of polycrystalline β-Mn was obtained by rapid quenching of 99.9% pure Mn into iced water from 900°C. A β-$Mn_{0.9}Al_{0.1}$ alloy was prepared by induction melting in Ar atmosphere using 99.9% pure Mn and 99.99% pure Al. The ingots were crushed into coarse powder for X-ray and neutron scattering measurements. No foreign phase was detected by X-ray diffraction. The temperature dependence of susceptibility was measured by using a torsion-type magnetic balance under an applied field of 8.23 kOe.

To determine uniquely the magnetic scattering contribution in these paramagnetic substances, polarized neutron scattering experiments were performed using the triple axis spectrometer, PONTA 5G, installed at JRR-3M of JAERI (Japan Atomic Energy Research Institute). Both constant energy (E) and constant wave vector (Q) scans were carried out at room temperature (290 K) and 7 K (and 60 K for β-$Mn_{0.9}Al_{0.1}$). The fixed incident neutron energy was 80 meV for pure β-Mn and 34 meV for the Al diluted sample and the collimations were chosen to be 40′-80′-80′-80′ and open-40′-80′-80′, respectively. The energy resolution of the spectrometer was $2\Gamma_{res}=19.3$ meV for the former and $2\Gamma_{res}=5.8$ meV for the latter conditions, as determined by vanadium incoherent elastic measurements. In order to calibrate the intensities and to measure the intrinsic polarization of the instrument, elastic scattering of the (221) nuclear Bragg peak was monitored under the various conditions used in the experiments. Typical flipping ratio of 18 ~ 20 was obtained under all conditions. Raw count rates for vertical ($H \perp Q$) and horizontal ($H /\!/ Q$) fields at the sample position and for flipper-off and -on conditions were corrected for finite polarization.[6] The subtraction of corrected spin flip counting rates of $H \perp Q$ from the $H /\!/ Q$ condition provides the pure magnetic scattering. These magnetic intensities are then corrected for the analyzer transmission in the manner of Dorner.[7]

Figure 1 shows the temperature dependence of magnetic susceptibility of pure β-Mn and β-$Mn_{0.9}Al_{0.1}$. The susceptibility of pure β-Mn is almost constant versus temperature, in agreement with a previous report.[8] For the β-$Mn_{0.9}Al_{0.1}$ alloy, the χ versus T curve exhibits a peak around 20 K, indicating formation of local moments and spin-glass freezing of the moments.

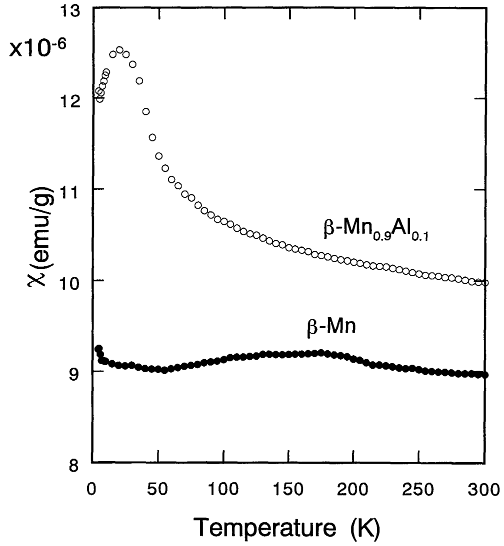

Fig. 1. Temperature dependence of the susceptibility of β-Mn and β-$Mn_{0.9}Al_{0.1}$ under an applied field of 8.23 kOe.

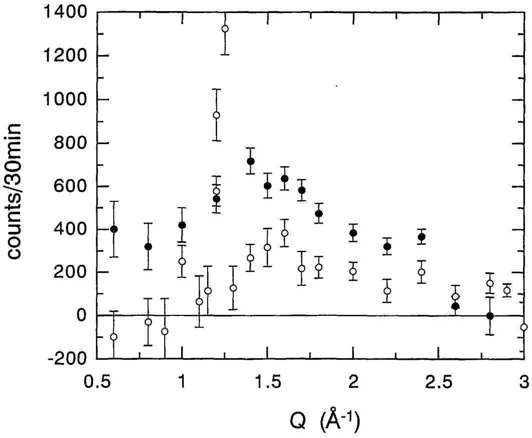

Fig. 2. Magnetic neutron scattering rate of β-Mn as a function of wave vector, Q, measured in the quasi-elastic mode ($\Delta E=0$) with energy resolution, $2\Gamma_{res}=19.3$ meV at 7 K (○) and 290 K (●).

Figure 2 shows the results of the quasi-elastic Q scan of β-Mn at room temperature and 7 K. The intensity is proportional to the integration of the squared amplitude of spin fluctuations over the energy within the energy resolution of 19.3 meV. A broad peak was observed at around $Q=1.5$ Å^{-1}. This wavelength corresponds to approximately twice the mean interatomic distance, indicating antiferromagnetic correlation of spin fluctuations. The amplitude decreases with decreasing temperature. However, it should be noted that scattering is still observed even at 7 K, suggesting the existence of strong zero point spin fluctuations, as discussed later. A sharp peak at $Q=1.2\,\text{Å}^{-1}$ of the spectrum at 7 K should be ascribed to the scattering associated to the (1/2 1/2 1/2) magnetic peak of antiferromagnetic MnO impurities, which is possibly introduced during experiments.

Figure 3 shows the results of energy scan of β-Mn for $Q=1.6\,\text{Å}^{-1}$ which almost corresponds to the peak position of the quasi-elastic Q scan. For a crude analysis of the results, we assume a simple Gaussian resolution function, $R(\omega)$ and a Lorenzian type scattering function. Then, the spectra were fitted to the following equations:

$$I(\omega)=C\int R(\omega-\omega')S(Q,\omega)\,\mathrm{d}\omega', \qquad (1)$$

where

$$R(\omega)=\frac{\sqrt{\ln 2}}{\sqrt{\pi}\,\Gamma_{\mathrm{res}}}\exp\left(-\ln 2\,\frac{\omega^2}{\Gamma_{\mathrm{res}}^2}\right), \qquad (2)$$

$$S(Q,\omega)=A(Q)\,\frac{1}{1-\exp\left(-\dfrac{\hbar\omega}{kT}\right)}\,\frac{\omega\Gamma}{\Gamma^2+\omega^2}. \qquad (3)$$

By choosing appropriate values of $C\cdot A(Q)$, and Γ, which are shown in Table I, we obtained fairly good fits, as shown in Fig. 3, where the dashed line indicates the deconvoluted spectrum, namely, $S(Q,\omega)$. At 7 K, scattering is observed only on the energy-loss side ($\Delta E>0$) of the spectrum. Since the spectrum spreads over a much wider range than the thermal energy of 7 K (≈ 0.6 meV), we may regard the magnetic scattering at 7 K as being caused by zero-point spin fluctuations as discussed by Takahashi and Moriya.[9]

Figure 4 shows the results of quasi-elastic scans of β-$\mathrm{Mn}_{0.9}\mathrm{Al}_{0.1}$ at 7 K, 60 K and 300 K. The profile of the spectra is similar to that for pure β-Mn, namely, it has a broad peak at around $Q=1.5\,\text{Å}^{-1}$, indicating the antiferromagnetic correlation between Mn spins. However, the scattering intensity increases with decreasing temperature, in contrast to pure β-Mn. Figure 5 shows the energy spectra for $Q=1.6\,\text{Å}^{-1}$ of β-$\mathrm{Mn}_{0.9}\mathrm{Al}_{0.1}$. The energy dependence of magnetic scattering is markedly different from that of pure β-Mn. The half-widths of the scattering function, which are given in Table I, are much narrower than that of pure β-Mn, particularly at low temperatures. These observations indicate that the spin fluctuations in the Al-substituted alloy become nearly static at low temperatures in ac-

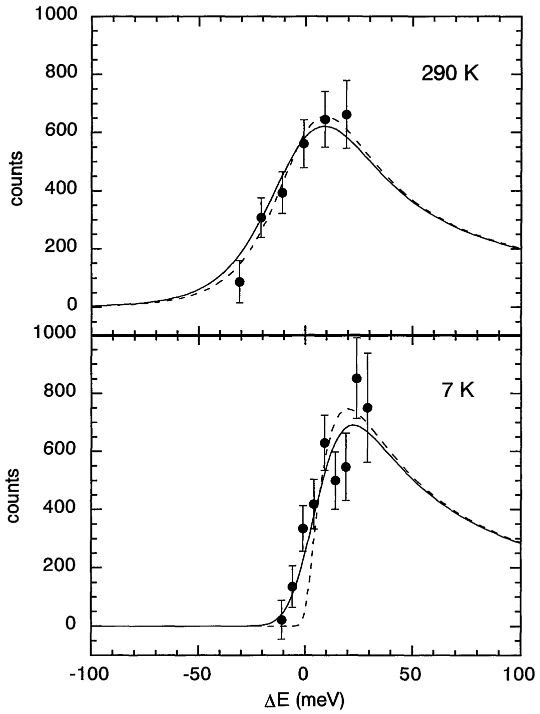

Fig. 3. Magnetic neutron scattering rate of β-Mn as a function of energy transfer, ΔE, measured for the momentum transfer, $Q=1.6\,\text{Å}^{-1}$ at 7 K and 290 K. The energy resolution is $2\Gamma_{\mathrm{res}}=19.3$ meV. Solid curve indicates the calculated spectrum using eq. (1) and parameters given in Table I. Dashed curve indicates the deconvoluted spectrum, i.e., $S(Q,\omega)$.

Table I. Parameters used for the fitting of the constant Q scan spectra at $Q=1.6\,\text{Å}^{-1}$ (Figs. 3 and 5) to eqs. (1)~(3).

	Temperature (K)	$2\Gamma_{\mathrm{res}}$ (meV)	$CA(Q)$ (Arbitrary units)	2Γ (meV)
β-Mn	7	19.3	1500±200	40±10
	290	19.3	720±100	60±15
β-$\mathrm{Mn}_{0.9}\mathrm{Al}_{0.1}$	7	5.8	1100±100	1.3±0.2
	60	5.8	200±20	4.5±0.5
	290	5.8	60±20	20±5

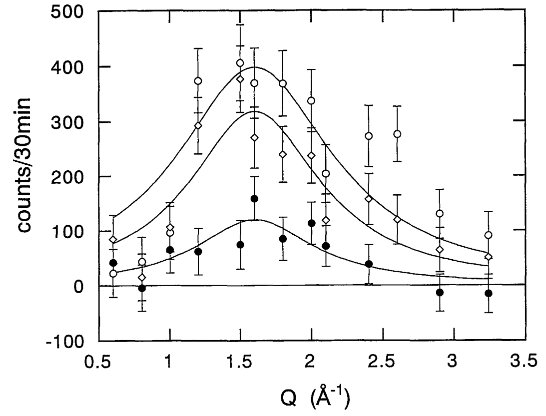

Fig. 4. Magnetic neutron scattering rate of β-$Mn_{0.9}Al_{0.1}$ as a function of wave vector, Q measured in quasi-elastic mode ($\Delta E=0$) with energy resolution, $2\Gamma_{res}=5.8$ meV at 7 K (○), 60 K (◇) and 290 K (●). Solid curves are the guide for eyes.

cordance with the spin-glass-like freezing at low temperatures. However, the difference in the energy spectra between 7 K and 60 K, which are located below and above the maximum in the χ-T curve, is not very appreciable, indicating that the dynamical spin correlation function does not change drastically between 7 K and 60 K in the time scale of neutron scattering. It is worth noting that very similar behavior was found in $Y(Mn_{0.9}Al_{0.1})_2$,[10] indicating the same mechanism for spin-glass freezing for both systems.

So far, we have not mentioned the absolute value of the amplitude of spin fluctuations. In principle, it is possible to estimate it by comparing the scattering intensity with a nuclear Bragg peak. However, in order to obtain the total amplitude, it is necessary to integrate the scattering intensity in both momentum and energy spaces. We have only estimated the equivalent squared Bohr magneton number of magnetic scattering within the energy range of 20 meV for the quasi-elastic spectra of both β-Mn and β-$Mn_{0.9}Al_{0.1}$ at 290 K. The results are shown in Fig. 6. The amplitude is much smaller than that of $Y_{0.97}Sc_{0.03}Mn_2$.[11] Noting, however, the narrower width of the energy window in the present measurements, the integrated intensity for the full energy range would give much larger intensity.

Noting the similarity in many physical properties of the $Y(Sc)(Mn_{1-x}Al_x)_2$ and β-$Mn_{1-x}Al_x$ systems, we consider that the dramatic change of magnetism by Al substitution for these systems can be understood in the same context as discussed in ref. 5. Namely, the ground state of the systems without Al impurity may be regarded to be in a quantum spin-liquid state, in the sense that there are strong quantum spin fluctuations with antiferromagnetic correlation. Such a state is possibly realized as a result of frustration in magnetic interactions. The substitution of nonmagnetic impurities releases the spin configurational degeneracy due to frustration and gives rise to a spin-glass state. Therefore, we

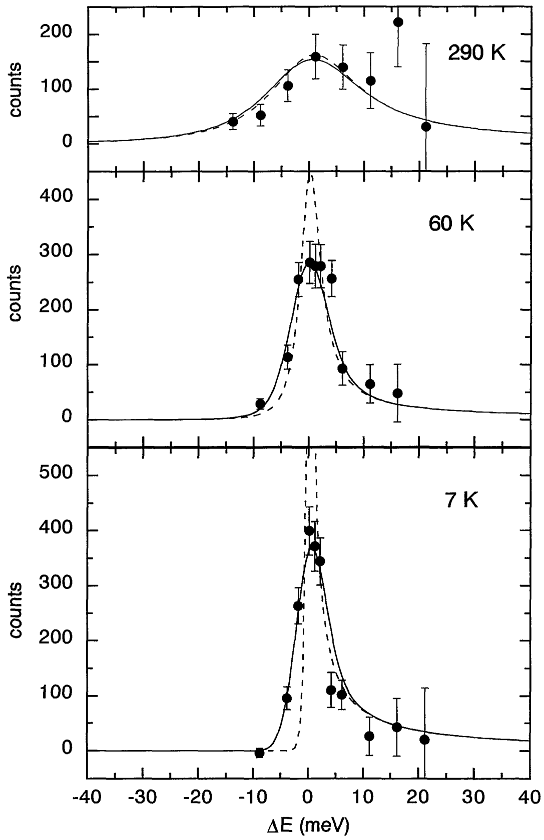

Fig. 5. Magnetic neutron scattering rate of β-$Mn_{0.9}Al_{0.1}$ as a function of energy transfer, ΔE, measured for the momentum transfer of $Q=1.6$ Å^{-1} at 7 K, 60 K and 290 K. The energy resolution is $\Gamma_{res}=5.8$ meV. Solid curve indicates the calculated spectrum using eq. (1) and parameters given in Table I. Dashed curve indicates the deconvoluted spectrum, i.e., $S(Q, \omega)$.

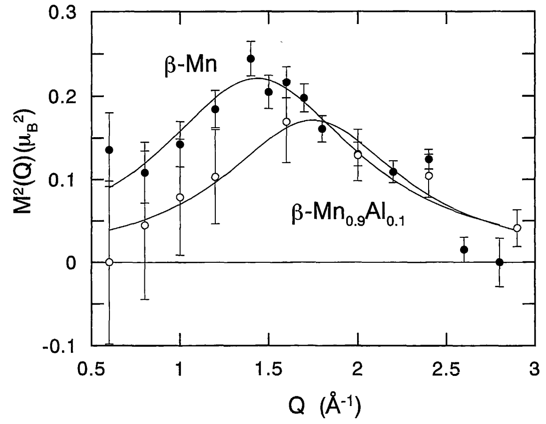

Fig. 6. Magnetic neutron scattering intensity of β-Mn (●) and β-$Mn_{0.9}Al_{0.1}$ (○) at 290 K as a function of wave vector, Q, measured in quasi-elastic mode ($\Delta E=0$) with energy resolution, $2\Gamma_{\text{res}}=19.3$ meV for both samples. The ordinate represents the equivalent squared Bohr magneton number of scattering amplitude.

interpret the present results of neutron scattering as follows. For pure β-Mn, the spin fluctuations at low temperatures are of quantum origin (zero-point fluctuations), which has rather high characteristic energy, and with increasing temperature, thermal fluctuations develop, resulting in an increase in total scattering. For β-$Mn_{0.9}Al_{0.1}$, the character of spin fluctuations is more static or of thermal origin. The frequency of fluctuations decreases with decreasing temperature, resulting in narrowing of the energy spectrum. Due to the narrow energy window used in this experiment, the slowing down of spin fluctuations gives rise to apparent increase of the intensity of magnetic scattering with decreasing temperature in the quasi-elastic spectrum. Details of analyses and discussion will be published elsewhere together with other experimental results, including NMR measurements.

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