# SANS Study of Slow Dynamics in Concentrated Spin Glasses

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In order to probe the microscopic mechanism of slow dynamics in spin-glasses, timeresolved small-angle neutron scattering (SANS) experiments on a concentrated spin glass alloy  $Fe_{65}(Ni_{0.866}Mn_{0.134})_{35}$  has been made. Time variation of the magnetic diffuse scattering pattern after being quenched into the reentrant spin-glass (RSG) phase persists up to 20 hours, as was observed in low-field susceptibility measurements. The time evolution of magnetic clusters in the RSG phase is discussed.

KEYWORDS: spin-glass, slow dynamics, SANS

## §1. Introduction

The problem of slow dynamics has long been a central issue of the research of spin-glasses. However, experimental studies of slow dynamics in spin-glasses have been limited to macroscopic measurements, such as the time evolution of thermo-remnant magnetization. In the previous neutron scattering studies on concentrated spinglass alloys we showed that magnetic clusters play an important role for the reentrant spin-glass (RSG) transition, and that a wide variety of spin dynamics observed by inelastic neutron scattering measurements can be attributed to the differences in the size and number of magnetic clusters.<sup>1-3</sup>) Based on these studies, we speculate that the mechanism of slow dynamics might be closely related to the time evolution of magnetic clusters which can be directly detected by small-angle neutron scattering (SANS) measurement.

We have made time-resolved SANS experiments on two kinds of concentrated spin glass alloys,  $Fe_{65}(Ni_{0.866}Mn_{0.134})_{35}$  and  $Cu_2(Mn_{0.70}Al_{0.30})Al$ , of which characteristics in the stationary state have already been clarified in the previous works.<sup>1–3)</sup>In this paper, we present a result of the former sample. A preliminary result of the latter sample has already been reported.<sup>4)</sup> It should be noted that the first observation of the time evolution of SANS pattern arising from the "clusters" was made in 1983 by one of the present authors and his co-workers.<sup>5)</sup>

## §2. Experimental Procedure

A single crystal of Fe<sub>65</sub> (Ni<sub>0.866</sub>Mn<sub>0.134</sub>)<sub>35</sub> alloy was grown by the Bridgman technique. Low-field magnetic susceptibility measurement showed that the alloy undergoes paramagnetic - ferromagnetic - RSG transitions. The transition temperatures are  $T_c=300$  K and  $T_{SG}=30$ K. SANS measurements were made utilizing the SANS-J spectrometer installed at the JRR-3M reactor of JAERI-Tokai. The incident-neutron wave-length and bandwidth are  $\lambda = 0.65$  nm and  $\Delta \lambda / \lambda = 10$  %, respectively. The momentum-transfer range of 0.03 < Q < 2 nm<sup>-1</sup> was covered with sample-detector lengths of 1.35 and 10 m. The sample was mounted in a closed-cycle refrigerator. External magnetic field was applied perpendicular to the incident neutron-beam direction so that the scattering intensities with the scattering vector parallel and perpendicular to the magnetic field are separately detected by a 2-dimensional detector. A sequence of time-resolved scattering pattern was taken with a typical time window of 10 min.



Fig.1. Relaxation rate of magnetization S(t) = dM(t)/dln(t) of Fe<sub>65</sub>(Ni<sub>0.866</sub>Mn<sub>0.134</sub>)<sub>35</sub> measured under various magnetic field. Measurements were made at T=6 K with  $t_w=1$  hr. Solid curves are drawn to guide the eye.



Fig.2. Neutron scattering intensity versus momentum transfer q from Fe<sub>65</sub>(Ni<sub>0.866</sub>Mn<sub>0.134</sub>)<sub>35</sub>. Solid and open symbols correspond to the total intensities measured between t=0-1 hr and t=15-16 hrs, respectively. Curves represent the fitting results described in the text.

#### §3. Results and Analysis

Various kinds of macroscopic measurements of slow dynamics have been made on the present sample.<sup>6)</sup> We show a typical example of long-time relaxation behavior of magnetization. The sample was rapidly cooled from ferromagnetic phase to T=6 K (RSG phase) in zero field. After a certain waiting time  $(t_w)$ , a magnetic field was applied at t=0 and the magnetization M(t)was measured as a function of time. Figure 1 shows the time evolution of the relaxation rate S(t) = dM(t)/dln(t)measured under various magnetic field for  $t_w = 1$ hr. The relaxation phenomena persists more than 20 hours and the maximum value of S(t) for the present experimental condition was observed under the field of 100 Oe. Based on the results of magnetization measurements, time evolution of SANS measurements were made following the similar procedure as above mentioned magnetization measurements. Figure 2 shows the neutron scattering intensity versus momentum transfer q. The measurement was made at T=8 K, H=120 Oe and  $t_w=30$ min. Solid and open symbols correspond to the total intensities measured between t=0-1 hr and t=15-16 hrs, respectively. In this figure only the data for q is perpendicular to H are shown. The result for the measurement with q is parallel to H is essentially same as the present one. The time variation of SANS pattern corresponds to the long-time relaxation process has been demonstrated by the present measurement. The observed SANS patterns are well traced by the sum of Lorentzian (LOR) and squared Lorentzian (SQL) functions in the momen-



Fig.3. Time dependence of the neutron scattering intensity from  $Fe_{65}(Ni_{0.866}Mn_{0.134})_{35}$  at representaqtive values of q. Measurements were made at  $T{=}8$  K and under the field of 120 Oe after  $t_w{=}1$  hr.

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$$I(q) = \frac{A_L}{q^2 + \kappa_L^2} + \frac{A_S}{(q^2 + \kappa_S^2)^2},$$
(3.1)

where  $A_L$  ( $A_S$ ) and  $\kappa_L$  ( $\kappa_S$ ) are the amplitude and the width (inverse correlation length) for the LOR (SQL) component, respectively. Solid and broken curves in Fig. 2 represent the fits to the data.

The next step is to determine the time evolution of these parameters and then to construct a real-space image of the time evolution of magnetic clusters. However, it has been found that we need additional measurements for larger q region for the precise determination of these parameters because a contribution of the LOR component is appreciable only for  $q > 0.2 \text{ nm}^{-1}$  region and the counting statistics was insufficient for this region. Alternatively, we show the time variations of the magnetic scattering intensity at representative q values. In Fig.3 magnetic neutron scattering intensities measured in a time window of 1hr are plotted as a function of time elapse after the application of the field. The left and the right columns represent the data for the measurements with q is parallel  $(I_{para.})$ , and perpendicular to  $H(I_{perp.})$ , respectively. The time variation persists up to 20 hours for all q range as was observed in the magnetization measurements. It should be noted that the relative increase of intensity with time elapse for q=0.20

 $nm^{-1}$  is considerably larger than those for smaller q values. Recalling the result of the previous works which showed that the LOR component of the magnetic diffuse scattering is attributed to the magnetic clusters,<sup>1-3)</sup> we conclude that the time evolution of magnetic clusters has been directly detected by the present measurement.

## §4. Conclusion

The present observation showed that the sensitivity of our experimental method satisfies the condition to detect the slow dynamics of RSG. A quantitative analysis including the determination of fitting parameters together with an improvement of the counting statistics by repetition of same heat cycles is under way.

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