Neutron Depolarization from Reentrant Spin Glasses

Yasuo ENDOH

Department of Physics, Tohoku University, Sendai 980

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The reentrant spin glass study was elucidated for the first time by means of the novel method of neutron depolarization. The wavelength dependent neutron depolarization senses the magnetic induction in mesoscopic scale inside the magnetic material. It was demonstrated that the domain distribution in the ferromagnet could be determined by the careful depolarization study of the magnetization process. This method has been applied to the reentrant spin glass problem, and the observation of the mesoscopic magnetic field gives a significant experimental fact to evince the reentrant transition from the ferromagnetic order. The quite recent experiments are reviewed in this paper, which contains many new results from various metallic reentrant spin glasses. The reentrant behavior is concluded to be governed by the degree of the frustration.

§ 1. Introduction

A new type of the ordered state of the "spin glass" is a well established concept, but there still exist many controversies in particular in classifying the real materials as the genuine spin glass.[1] The competition of the exchange interaction which introduces the frustration and the random distribution of either magnetic species or magnetic bonds are a necessary ingredient for the realization of the 'spin glass'. Any real spin glass materials naturally form either magnetic alloys or random mixtures of more than two components of at least one magnetic element, which potentially contain dirts. Since the singularities in thermodynamic properties associated with the spin glass transition are usually not so strongly enhanced, the background noise from such dirts may overwhelm the magnetic signal associated with the transitions. Furthermore even if an ideal spin glass material is obtained, many experimental results are history dependent, which makes a universal determination difficult. In this respect, the experimental results on the spin glass subjects should be presented with the clear documents on the history in the experiment, otherwise unnecessary controversial arguments may be encountered. In some cases, completely opposite interpretations may exist for exactly the same experimental result. Nevertheless it is certainly a hard task to extract commonly acceptable conclusions not only due to the above mentioned experimental difficulties but due to the fact that all the experimental evidences are not always consistent. In some cases any singularity in thermodynamical properties is not observed, which should appear at the transition in a homogeneous system: For instance, the well defined cusp appears in the susceptibility at a certain temperature, but no appreciable anomaly appears in the specific heat at the same temperature.

Furthermore, the mean field theory predicts a well defined single order parameter to characterize the 'spin glass' phase.[2] It does not always contain such a single order parameter in the spin glass systems, because it is not ergodic. It means that the mean field approximation may be far simpler than the result of the precise treatment of thermodynamics in the spin glass. Even though it contains a weakness, the mean field theory provides many useful predictions like the Gabay-Toulouse[3] or the Almeida-Thouless line[4] in the context of the Sherrington-Kirkpatrick model.[5]

The reentrant spin glass (RSG), which is the main issue in this paper, is also given by the concept of the mean field theory. It was defined to the sequential transition with decreasing temperature; the system first undergoes a continuous transition from paramagnetic to the long range ordered (LRO) phase at the Curie temperature, T_c and then at the lower temperature, usually called the glass temperature, T_f, it reenters into a magnetic phase which resembles the spin glass state. Again many controversies exist on this phase transition from both experimental and theoretical points of views.[6]

At the present stage, a commonly observed 'bell' shape of thermal evolution in the low field magnetization is recognized to be a unique feature to characterize the RSG. The transition temperature T_f drastically shifts with the strength of the applied field, even in the low field region. The fact clearly indicates that the spin glass transition may be triggered by the small molecular field involving large numbers of spins. Thus the magnetic behavior near T_f in the spin glass systems is essentially different from the critical behavior near the continuous phase transition of the second order, where the correlation length of the order parameter developes continuously from the short distance in atomic scale to the infinitely long distance. On the other hand, in the spin glass transition, the singularity in the spin correlations may only be appreciable at the longer length scale with tiny or nonvisible change in thermal properties of the short range spin correlations.

Therefore it is very important to detect any magnetic anomalies in the larger scale at the phase transition, which may be an order of the ferromagnetic domain size. This scale is defined as the mesoscopic scale, which is typically between several orders of nm and sub μm. Small angle neutron scattering or quasielastic magnetic scattering detecting such an anomaly in the large scale is an orthodox method. However this method encounters an essential difficulty to substract non-magnetic background which usually overwhelm the magnetic signal. Light scattering or electron diffraction method is also capable, but the experiments are not always easy for metallic magnets which are not optically transparent. Then the neutron depolarization provides another excellent tool for the study of the magnetism of the spin glass by detecting the spatial magnetic density fluctuations in mesoscopic scale as will be described in the following section.

We now focus on the present theoretical comprehension on the RSG subject. In the mean field approximation,[2] the onset of the RSG phase or the disappearance of the ferromagnetic LRO is defined to result from the replica symmetry breaking or the freezing of the transverse component of the magnetization which destroys the complete ordering of the single component LRO, longitudinal to the average magnetization. Therefore the RSG is not a reentrance in the thermodynamical point of view,

but the mean field theory predicted that the RSG state is characterized by the coexistence of the spin glass like disordered state superposing the ferromagnetic LRO states.[3] In other words, it consists of the randomly oriented frozen transverse component and the long range ordered longitudinal component below T_f, which can be regarded as the mixed state. Then there exist many controversies on the phase transition in view of thermodynamics as well as the symmetry argument of the system, which may differ from the above mentioned mean field concept.[7]

In this review paper, we describe the recent experimental studies on the RSG subject by using many RSG alloys by means of the novel neutron depolarization method,[8] and we intend to discuss our view of the RSG summarizing the results.

§ 2. Neutron depolarization as a function of neutron wavelength

The neutron depolarization measurement is a simple method to study the spatial inhomogeneity of the magnetic field inside a bulk magnet by detecting the change of polarization of the transmitted neutron beam through the magnetic materials. In 1941, Halpern and Holstein (H, H)[9] theoretically derived how neutrons are depolarized by the magnetic induction in a ferromagnet consisting of multidomains. Then a new experimental method to measure the wavelength dependent depolarization was proposed to provide rich informations on the domain structure in the ferromagnets. Although depolarization experiments to study the magnetization processes have been initiated a long time ago, since then, there was no wavelength dependent experiment until the recent years.[10] The first wavelength dependent measurement utilizing polychromatic pulsed polarized neutrons demonstrates that the wavelength dependence of neutrons transmitted through ferromagnets reflects the domain orientation in the magnetization process just as predicted in the H, H derivation.[11] Furthermore, the domain size can be readily determined by the wavelength dependent depolarization data.

The principle of H, H derivation is described in some detail. The neutron polarization, $\boldsymbol{P}$ is treated as a classical vector spin, which follows a classical equation of motion under the stational magnetic field,

$$d\boldsymbol{P}/dt=\gamma\boldsymbol{P}\times\boldsymbol{B}(\boldsymbol{r})\,. \tag{1}$$

The validity of this principle is already confirmed by the fact that the Mezei spin flipper perfectly works,[12] which is a realization of this principle. In other words, when polarized neutrons travel through the magnetic field applied in perpendicular direction with respect to the neutron polarization, the neutron polarization is flipped over at the condition of $DH=\pi/C\lambda$ after the Larmor precession around the field. The product of DH represents the integrated field over the neutron path length. C is the physical constant related to the nuclear gyromagnetic ratio of the neutron spin. The measurable polarization in the depolarization experiments is therefore the integrated value of Eq. (1) over the transmitted neutron path length D of the ferromagnetic slab. In order to determine $\boldsymbol{H}(r)$ even in a simple case of the uniform field, the data of the wavelength (λ) dependence is necessary. Nevertheless, this treatment is within the

classical limit where the change in the magnetic field occurs gradually at the longer distance than the neutron wavelength, λ. Therefore this depolarization method of transmitted neutrons is not sensitive to the short magnetic fluctuations in an atomic scale. It was also confirmed that no depolarization occurs in a usual paramagnet or even some spin glass materials, in which spins fluctuate fast in time and also do spatially in the short distance.

In principle, the three dimensional vector analysis of neutron depolarization can be performed on the TOP spectrometer installed at the pulsed neutron scattering facility of the National Laboratory for High Energy Physics (KEK).[13] The experiments so far done, however, by using the spin glass materials are the polarization analysis of one component parallel to the initial neutron polarization. Then the polarization P_f of transmitted beam is readily interpreted by the integration of Eq. (1).

Let us describe the neutron depolarization for the ferromagnetic bulk material, which is a simpler case than the reentrant spin glass case of our main concern in this paper. Usually a thin ellipsoidal plate is used in the depolarization experiment in order to evaluate the demagnetization effect easily. P_f is given by the following equation; note that P_f is now taken only the projection along z direction,

$$P_f(\lambda)=[1-\langle(1-\langle\cos\{CB\delta\lambda\}\rangle_\delta(B_x^2+B_y^2)/B^2\rangle_B]^{D/\delta}. \tag{2}$$

Here $\langle\cdots\rangle_{B,\delta}$ represents the ensemble average over either the local magnetic induction $\mathbb{B}$ in each domain or the domain size δ. D and δ are respectively the sample thickness or the neutron path length and the average domain size along the neutron path direction. λ is the neutron wavelength, and C is the physical constant and is given to be 4.63×10^{-2} in $\mathrm{cm^{-1}Oe^{-1}\AA^{-1}}$ units. This equation (2) just represents that the neutron depolarization occurs by the vector spin rotation around magnetic induction $\mathbb{B}$ in each domain. The resultant polarization of the beam after travelling through the ferromagnetic slab is therefore the multiplied values of the depolarization of an independent event in each domain, which is N times; $N=D/\delta$. Once the averaging procedure is done through the proper approximation, P_f is given as a function of neutron wavelength λ.

Although expressions for the general case are not simple, functional forms are available in several limiting cases of the ferromagnets consisting of either small or large domains. For the former case of the small domains, the Larmor precession of neutrons in each domain is far smaller than the complete turn in each domain or $CB\delta\lambda \ll 2\pi$. Then P_f is given as follows:

$$P(\lambda)=[1-\langle1-\cos\{CB\delta\lambda\}\exp(-\alpha\lambda^2)(B_x^2+B_y^2)/B^2\rangle_B]^N. \tag{3}$$

A slightly complicated form, which is approximated by the single exponent of λ, $\exp\{-\beta\lambda\}$, is also obtained for the case of the comparatively larger domain case, and thus either $\exp\{-\beta\lambda\}$ or $\exp\{-\alpha\lambda^2\}$ should be seen for the small domain limit, since the cosine term usually becomes a constant value against λ after the average over $\mathbb{B}$.[11] In fact we have seen both wavelength dependent features as shown in the next section. On the other hand, for the latter case, where neutrons precess many turns in each domain, P_f is constant with respect to λ. Neutrons are completely depolarized if the

domains are randomly oriented where the external field is less than the demagnetizing field,

$$P_f(\lambda)=[1-\langle(B_x^2+B_y^2)/B^2\rangle_B]^N . \tag{4}$$

In some cases, however, upon reaching a technical magnetization stage, the oscillatory variation is seen in the wavelength dependent polarization. The fact depends on the magnetic domain structure; for instance, if the domains penetrate through from one side to the other in the thin plate of the sample, the polarization of transmitted beam through such a ferromagnetic plate shows a cosine oscillation, because travelling neutrons rotate around the uniform magnetic induction in the penetrating domain, or $N=1$. In the real experiment, however, the beam cross section is usually wider than the domain size. Then the averaging procedure is still necessary across the beam crosssection, because there are a large number of domains in the plate. Then the polarization P_f is approximated in the following functional forms, which was initially obtained from our previous experiments:[11]

$$P_f(\lambda)=P_0+(1-P_0)\exp(-\sigma_I\lambda^2)\cos(I\lambda) , \tag{5}$$

$$I=CB\delta\Sigma , \quad \delta\Sigma=D , \tag{6}$$

$$\sqrt{2\sigma_I}/I=\Delta D/D=\text{const} . \tag{7}$$

We previously demonstrated two limiting cases of ferromagnets with large and small domains and succeeded in determining the domain size by using the wavelength dependent depolarization data. We used the same ferromagnetic alloy of $Fe_{0.85}Cr_{0.15}$ with the different heat treatment.[11] The data from annealed sample are essentially independent of the wavelength of neutrons, and however they show the exponential decay in polarization with respect to the wavelength for the quenched sample with the small domains as anticipated. The data are shown in Fig. 1, which were analyzed by using the standard expressions either for the case $CB\delta\lambda\gg 2\pi$ or $CB\delta\lambda\ll 2\pi$. As the result, we could determine two parameters of their domain size and orientation with respect to the field. The domain size of these examples is about 200 μm and 3 μm for the annealed and quenched alloys respectively. Therefore $D/\delta=N$ is either 5 or 500 for two cases. In this analysis we fixed the magnetic induction value as obtained by the sum of the external field and 4π times the saturation magnetization, $B=H_{ex}+4\pi M$, and eventually found it to be a reasonable postulate. It must be emphasized that the depolarization is still apparent in a certain external field where the magnetization curve nearly reaches the saturated value. This fact is not unusual because the depolarization occurs by the rotation around the field component perpendicular to neutron polarization. For the case of the small domains where D/δ goes to the large value, the depolarization becomes large due to many nonadiabatic processes at each domain boundary during the beam passage through the sample and eventually final polarization P_f becomes smaller than unity. On the other hand, the bulk magnetization measurements give a saturated value. This example evidently shows that the depolarization is a unique probe to sense the spatial distribution of the magnetic induction perpendicular to the average magnetization direction. In particu-

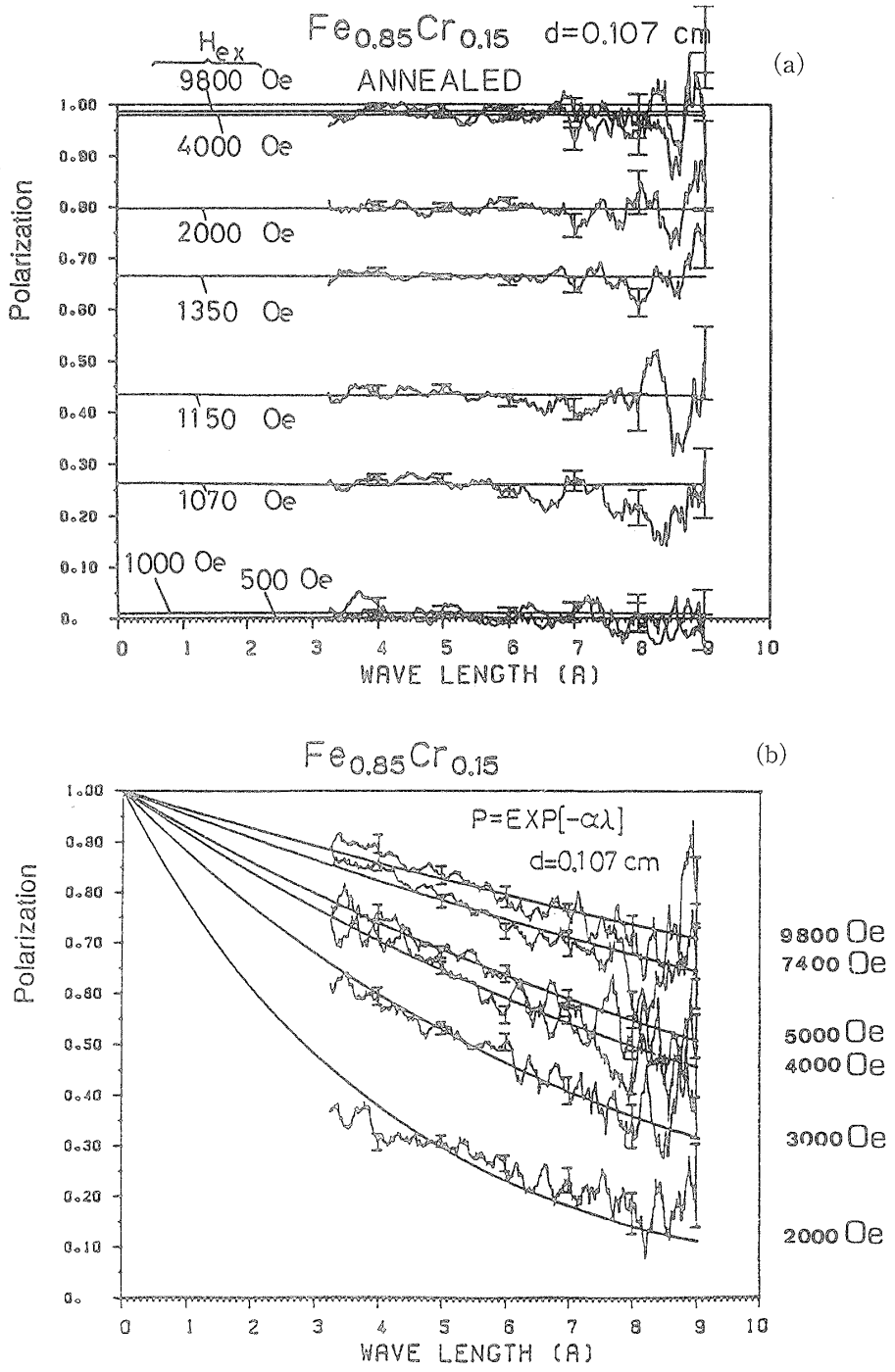


Fig. 1. Wavelength dependent polarization $P(\lambda)$ of transmitted neutrons through polycrystalline ferromagnetic plates of 0.107 cm thick. (a) Annealed and (b) quenched $Fe_{0.85}Cr_{0.15}$ were measured in the different external field at room temperature.

lar for the study of the reentrant spin glass subject, it is very important to detect the spatial magnetic density fluctuations in mesoscopic scale and also the reentrance is predicted to appear by freezing of the transverse component of the magnetization. Therefore the neutron depolarization method stands as the powerful tool for the reentrant spin glass properties, and furthermore it is emphasized that the experiment is very simple as described in the following section.

§ 3. Reentrant spin glass alloys

We again emphasize here that the reentrance is defined as thermal phenomenon that, on cooling the RSG material, it passes from paramagnetic to another magnetic state with no spontaneous magnetization through the long range ordered phase. Of course, the ordered phase includes not only ferromagnetic but antiferro- or helimagnetic phases, but in the present experiment only the ferromagnetic case was treated due to the fact that the appreciable depolarization can be expected in the ferromagnetic RSG. As is shown in Fig. 2, thermal evolution of the low field magnetization from the ferromagnetic RSGs commonly shows a "bell" shape due to the disappearance of the spontaneous magnetization below T_f. It is also common that the RSG appears in the phase diagram spanned on the x (concentration)-T(temperature) plane near the critical concentration where a T_c line quickly approaches $T=0$, and a new phase boundary line of T_f turns into the LRO phase as depicted in Fig. 3. Then a question is whether this phase below T_f is a replica symmetry breaking state. In other words, whether these lines represent the phase boundary as Parisi suggested.[14]

In this section, typical data of the wavelength dependent depolarization measurement are displayed on the sequence from the weakly frustrated RSG to the strong frustrations. $Au_{1-x}Fe_x$ alloys are categolized as the typically weak frustration RSGs.[15] The wavelength dependence for the $Au_{0.81}Fe_{0.19}$ is shown in Fig. 4. The

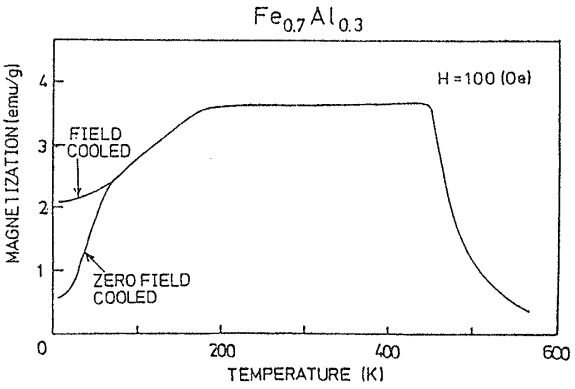


Fig. 2. Thermal evolution of magnetization from $Fe_{0.7}Al_{0.3}$ RSG alloy. The "bell" shape of the magnetization curve at low magnetic field (100 Oe) is shown here as a typical character of RSG. Below 100 K irreversibility is observed.

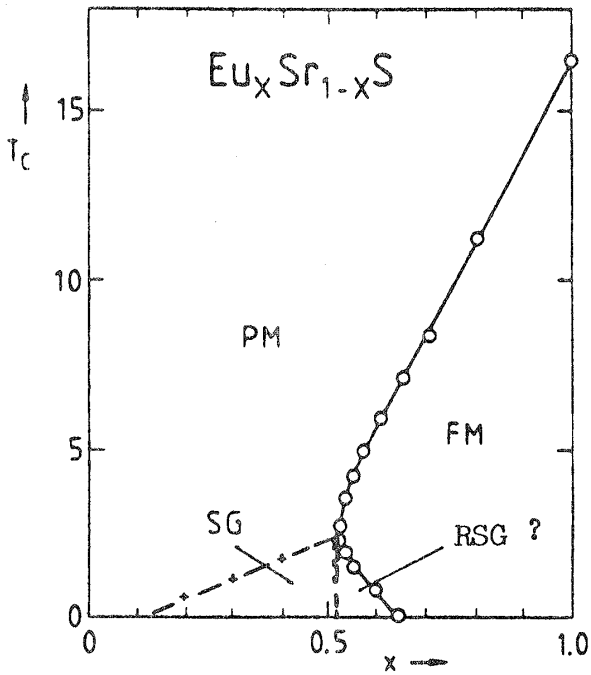


Fig. 3. Typical example of the magnetic phase diagram of the reentrant spin glass system of $Eu_xSr_{1-x}S$. PM, FM, SG and RSG represent respectively, paramagnetic, ferromagnetic, spin glass and reentrant spin glass states.

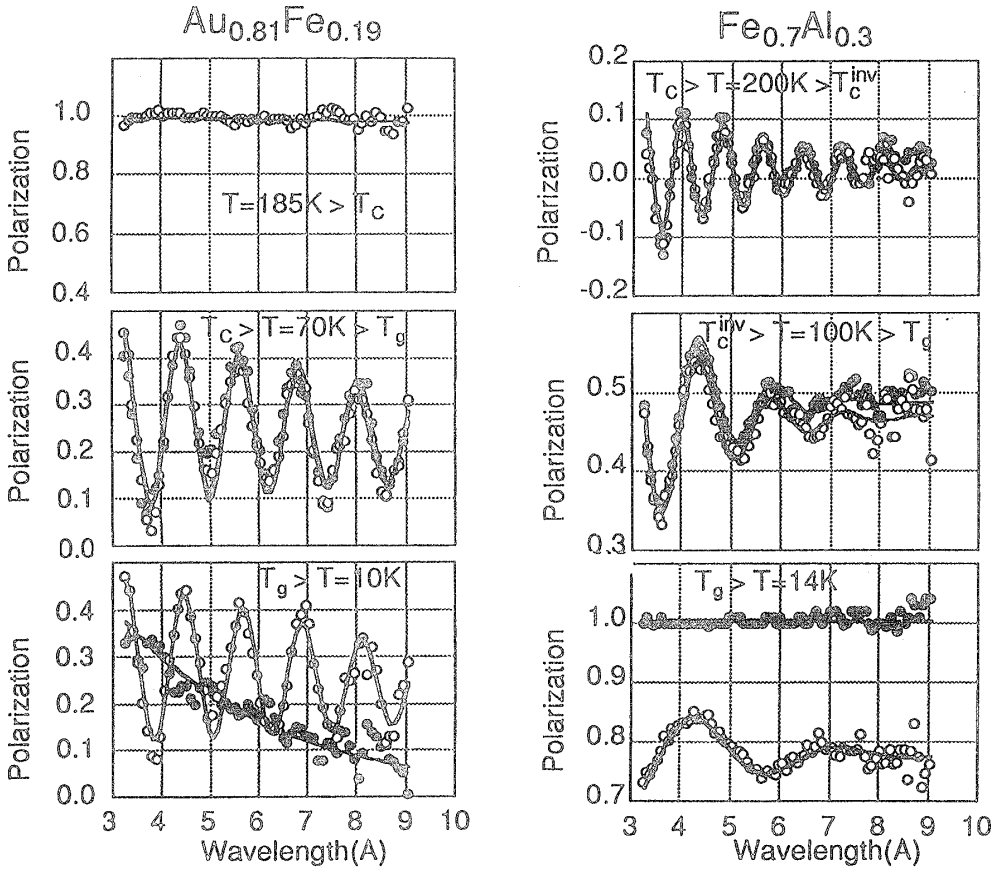


Fig. 4. Wavelength dependence of polarization $P(\lambda)$ from $Au_{0.81}Fe_{0.19}$ alloy is shown in the left hand row in three different phases; paramagnetic (top), ferromagnetic (middle) and RSG state (bottom). $P(\lambda)$ from $Fe_{0.7}Al_{0.3}$ RSG alloy is in the right hand row; ferromagnetic above T_c^{inv} (top), ferromagnetic below T_c^{inv} (middle) and RSG state (bottom). Filled and open data points respectively correspond to ZFC and FC measurements.

depolarization results from $Fe_{0.7}Al_{0.3}$ are also presented in the same figure as the comparison. It is remarkable that the qualitatively different depolarization curves of λ dependence are visible in the three different phases in zero field cooling (ZFC). Namely no depolarization occurs in the paramagnetic phase above T_c and the sinusoidal oscillatory λ dependence in the polarization is seen in the intermediate phase between T_c and T_f. Finally it changes to the exponential decay in the λ dependent polarization below T_f. On the other hand, at the field cooling (FC) process, essentially no appreciable temperature dependence occurs in these depolarization curves below T_c. In this system of $Au_{1-x}Fe_x$ RSGs, the oscillatory λ dependence is always seen in the intermediate ferromagnetic phase as well as below T_f in the FC samples. The fact evidentally indicates that the magnetic domains in the ferromagnetic state penetrate through one side to the other in the disk samples about a half mm thick, which is clear from the description in the preceding section.

The wavelength dependent curves in the polarization could be analyzed by adopting the depolarization function of the multidomained ferromagnet, and the parameters required to the RSG studies have been extracted. In particular, the important parameters of both the magnetic induction I^* as well as the domain size δ could be obtained unambiguously from the sinusoidally oscillatory curves.

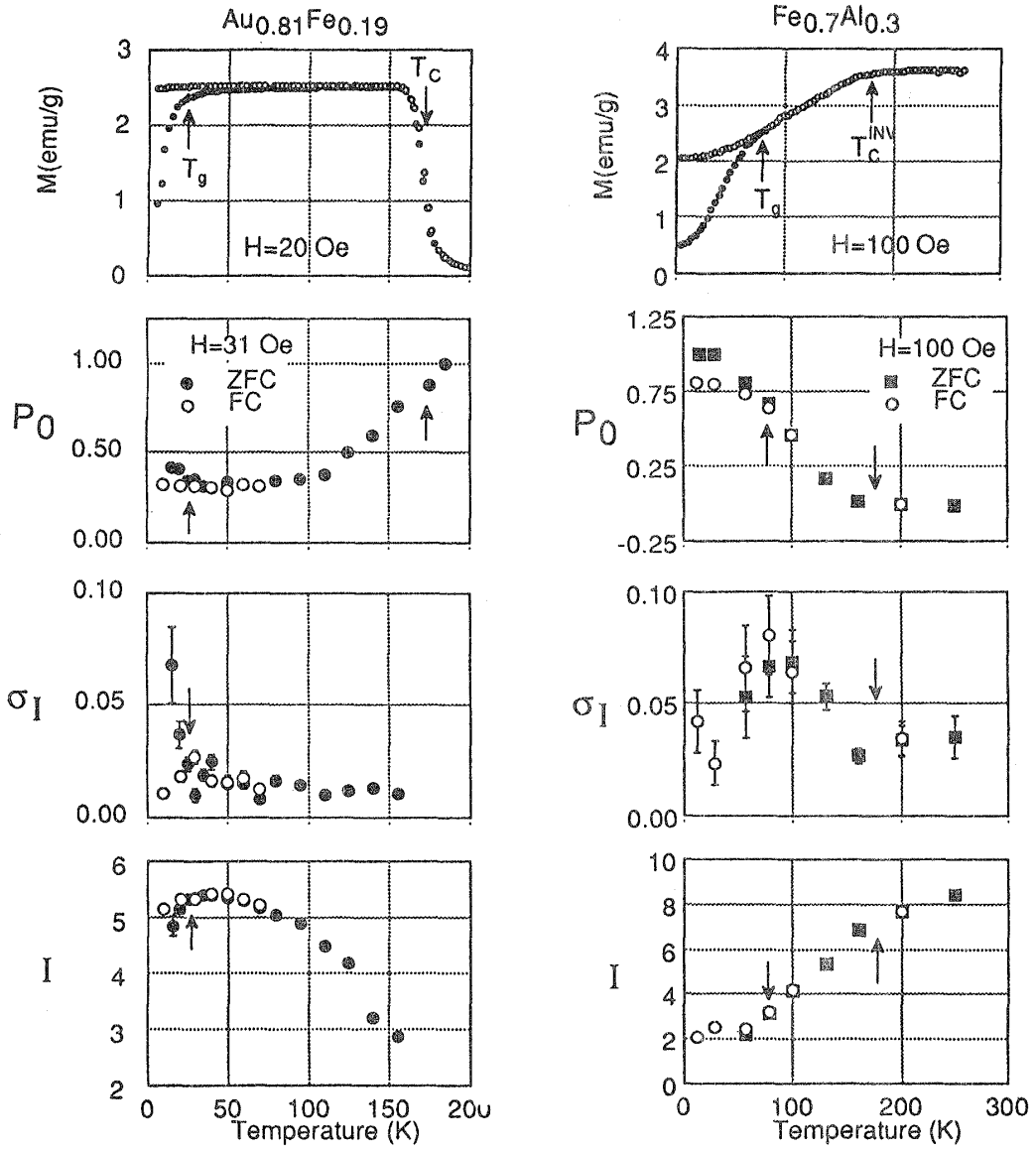
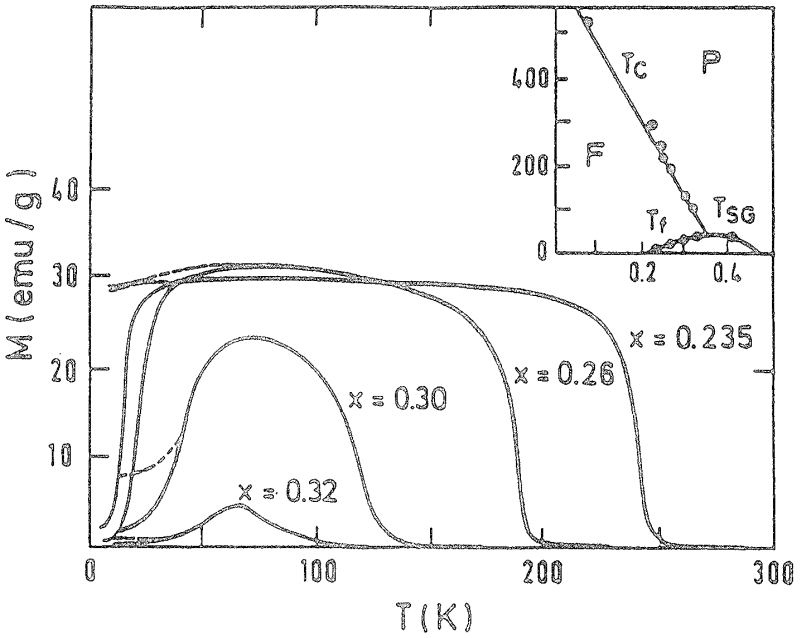
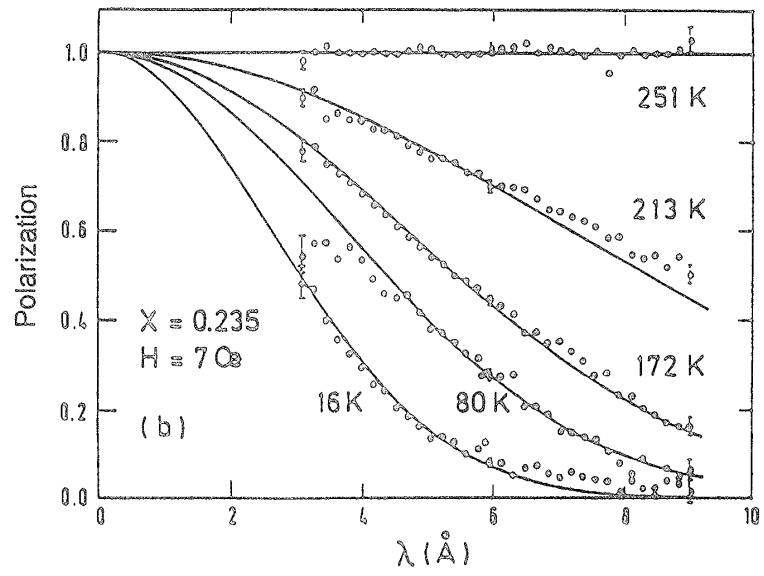


Fig. 5. Temperature dependence of the parameters of P_0, σ_I and I^* in Eq. (5) of the fitted function for the depolarization data together with the low field magnetization curves of $Au_{0.81}Fe_{0.19}$ RSG alloy (left hand row) and $Fe_{0.7}Al_{0.3}$ RSG alloy (right-hand row).

When we look at the thermal evolution of I^* in the AuFe RSG which is shown in Fig. 5, we immediately find the fact that I^* is less than the saturation magnetization value which decreases below T_f. No extra domain distribution with different sizes below T_f could be detected in this RSG alloys. Note that the δ value determined just below T_f is constant with temperature in the ferromagnetic phase. Then I^* was found to decrease with the further decrease of temperature when δ is assumed to be fixed. This important result tells us that the spontaneous bulk magnetization averaged over all atoms in each domain, which is proportional to I^*, is remarkably reduced at low temperatures even above T_f. The mixing of the secondary disordered phase in mesoscopic scale is not expected in the RSG phase, since I^* is uniform even at the lowest temperature well below T_f. To summarize here, the magnetization averaged over each domain considerably decreases in the RSG phase in the $Au_{1-x}Fe_x$ RSG alloys,[16] which was observed for the first time in the real experiment.



(a)



(b)

Fig. 6. Thermal evolution of magnetization curve in amorphous $Fe_{1-x}Mn_x$ alloys of $x=0.235$, 0.26, 0.30 and 0.32 with magnetic phase diagram (insert) in (a). $P(\lambda)$ of amorphous $Fe_{0.765}Mn_{0.235}$ RSG alloy at different temperatures of 16, 80, 172, 213 and 251 K are presented in (b).

The next examples are the amorphous RSGs of both $Fe_{1-x}Mn_x$[17] and $Ni_{1-x}Mn_x$ alloys[18] containing some metalloid elements such as B, N, etc., for the stabilization of the amorphous phase. The RSG state appears around $0.2<x<0.3$, where T_c asymptotically approaches zero in both amorphous alloys. The depolarization data of the exponential form with respect to the wavelength illustrated in Fig. 6 are very similar to the results of the ferromagnet with small domains. It turns out however from the analysis combined with the magnetization data shown in Fig. 6 that the depolarization can be determined solely by the magnetization or the magnetic induc-

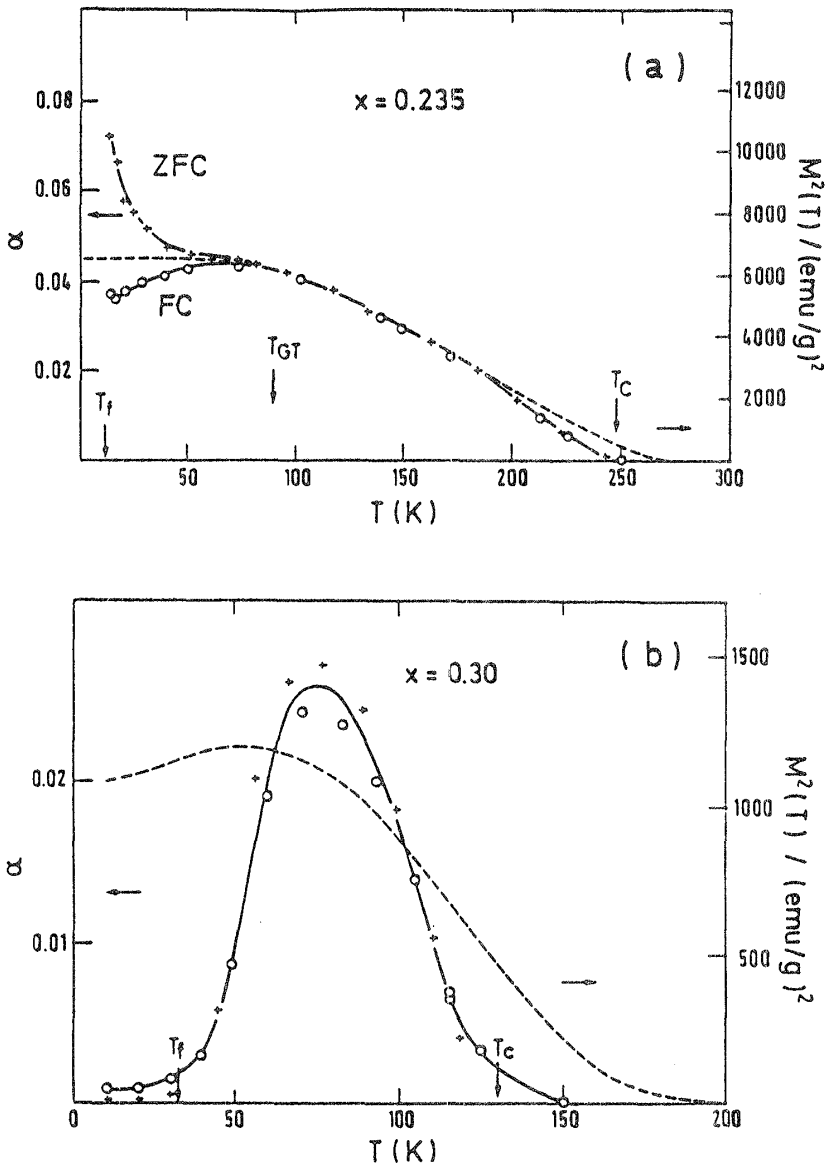


Fig. 7. Temperature dependence of the coefficient a of the exponential curve of $P(\lambda)$ for amorphous $Fe_{0.765}Mn_{0.235}$ RSG (a) and $Fe_{0.7}Mn_{0.3}$ RSG (b) alloys together with the $M^2(T)$ curves.

tion in the domain, and that the domain size is temperature independent similar to the result from $Au_{1-x}Fe_x$ glasses. The results from both amorphous FeMn add NiMn alloys are essentially similar except the details. The irreversible thermal evolutions of the amorphous $Fe_{0.765}Mn_{0.235}$ alloy are appreciable at low temperatures where the magnetic susceptibility drops substantially.

Another significant result is the appearance of the minimum in the thermal evolution of the polarization at which the magnetization reaches maximum in the amorphous $Fe_{0.7}Mn_{0.3}$ alloy. In other words, toward the lowest temperature below the reentrant transition, the polarization recovers as observed typically in the $Fe_{0.7}Al_{0.3}$ RSG.[18] In this respect it is noted that entirely different features of thermal evolution of the depolarization were observed in two samples of amorphous $Fe_{0.765}Mn_{0.235}$ and $Fe_{0.7}Mn_{0.3}$ alloys. This reflects that the coefficient of the gaussian function of the polarization is nearly proportional to the square of the magnetization in the $Fe_{0.765}Mn_{0.235}$ alloy, which is shown in Fig. 7(a). As compared with Fig. 7(b),

this result shows a significant contrast.

The common feature, on the other hand, is that the polarization increases with the increase of temperature as a whole. The data were successfully analyzed to be the temperature independent domain size and the coefficient of the exponential form of λ dependence in polarization is approximately proportional to the square of the magnetization as seen in Fig. 7(a). This experimental result is well interpreted by adopting the mean field approximation such that freezing of the transverse component with respect to the average longitudinal magnetization direction induces the blocking of the domain wall motion, which gives the reduction of both mesoscopic field and the average atomic moment value in the RSG phase. Since the depolarization occurs only by the longitudinal magnetization, the average induction in the domain should vary with temperature like the magnetization measured at technical saturation, whereas the transverse component has little effect on the depolarization due to the short correlation length compared with the domain size. In the mean field theory this magnetic state is defined to be the canting state, which might have little influence on the polarization but be attributed to the appearance of an additional anisotropy like Dzaloshinskii-Moriya interaction, which prevents free domain wall motion. Therefore the appreciable history dependent behavior in the depolarization as well as magnetization measurements appears below T_f, being dependent on the cooling processes. The experimental data show the persistence of domains at low temperatures below the reentrance in the weakly frustrated RSGs, where the bulk magnetization drops substantially. The data analysis suggests the decrease of the domain size around 5 μm, where the average magnetization of each domain is randomly oriented at low external field, but, on the other hand, above T_f the domain size substantially

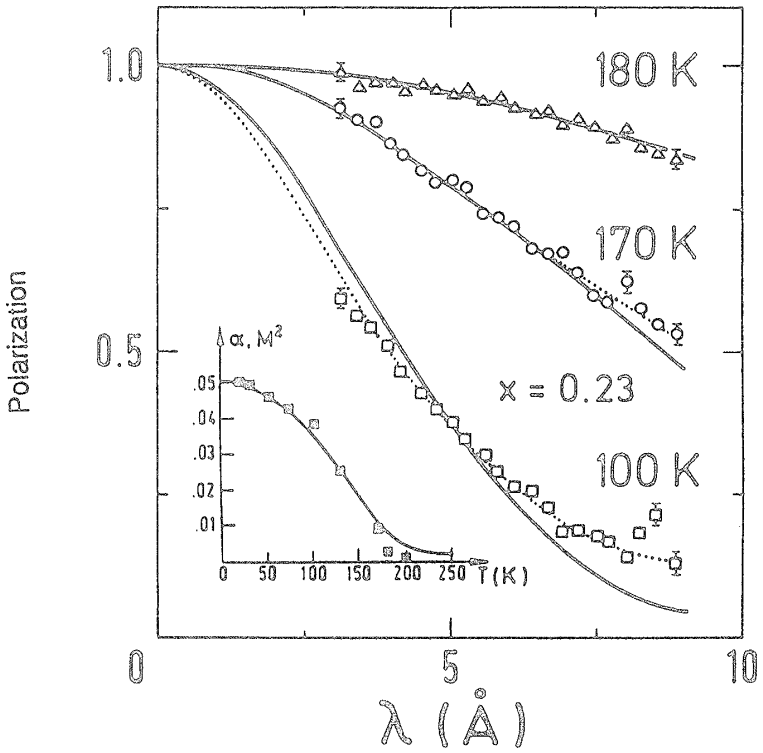


Fig. 8. $P(\lambda)$ of amorphous $Ni_{0.77}Mn_{0.23}$ alloy at several temperatures of 100, 170 and 180 K, and temperature dependence of α for the exponential λ dependent depolarization data.

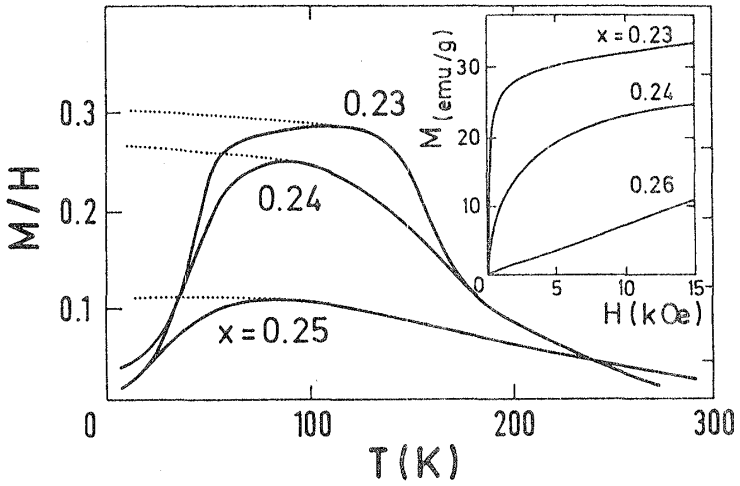


Fig. 9. Magnetization data for amorphous $Ni_{1-x}Mn_x$ alloy with $x=0.23$, 0.24 and 0.25. M/H is shown as the function of temperature, which was obtained from the initial slope of the magnetization curves inserted as typical examples at 100 K. Thick lines are initial slope while dotted lines are reversed slopes.

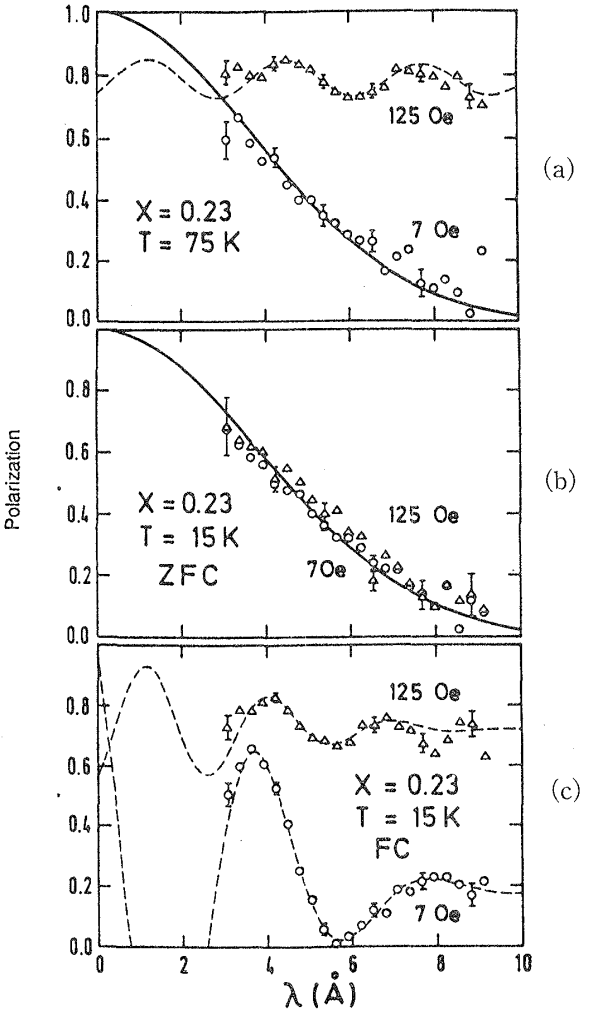


Fig. 10. $P(\lambda)$ for $Ni_{0.77}Mn_{0.23}$ alloy in low field (7 Oe) and high field (125 Oe). (a) $T=75$ K (b) $T=15$ K with ZFC and (c) with FC.

increases and the orientation of the magnetization is nearly parallel to the external field.

As for amorphous $Ni_{1-x}Mn_x$ alloys, the depolarization data are essentially similar to the $Fe_{1-x}Mn_x$ data as shown in Fig. 8. Though the temperature dependence of the magnetization evolves the bell shape due to the substantial decrease below T_f as shown in Fig. 9, the mesoscopic magnetic induction detected by the depolarization measurement is saturated at low temperatures. Considerable history dependent behaviors with ZFC and FC processes were also observed. The history dependent feature detected by the depolarization method is much clearer than any other magnetic measurements as shown in Fig. 10.

Finally the experimental results from strongly frustrated RSG systems are presented. The most typical example is $Fe_{0.7}Al_{0.3}$ RSG.[15] In order to comprehend the qualitative difference in depolarization results, important data are shown in Figs. 4 and 5. The typical bell shape magnetization curve similar

to the weakly frustrated RSGs are seen in this material as shown in Figs. 2 and 5(e). However thermal evolution of depolarization particularly below T_f is entirely different from that of the previous results from the weakly frustrated RSGs as is compared in Fig. 4. As shown in Fig. 4, no depolarization occurs in both the paramagnetic above T_c and the reentrant phase below T_f. If we interpret this fact of no depolarization below T_f as the evidence of the complete loss of the spontaneous magnetization or the disappearance of the ferromagnetic LRO, the magnetic state in the $Fe_{0.7}Al_{0.3}$ RSG below T_f should be completely different from that of the $Au_{0.81}Fe_{0.19}$ RSG. This qualitative difference in the experimental results is the first clear evidence suggesting that there exist different RSG states, even though the low field magnetization looks similar like a common bell shape.

In the ferromagnetic phase above T_f, the observed curve is characterized by the damped oscillatory λ dependence suggesting that the sample is in a large domain state, which penetrates one side to the other. Looking at the data above T_f of the damped oscillatory λ dependence, we parametrized the λ dependent polarization $P(\lambda)$ by fitting Eq. (4) to the experimental curves as shown in Fig. 5. Since I^* was found to decrease with decreasing temperature towards T_f, and furthermore it continuously decreases below T_f at lower temperatures, which is much more dramatic than the thermal evolution of the AuFe RSG. Another characteristics in $Fe_{0.7}Al_{0.3}$ RSG is the large damping factor in the wavelength dependent polarization, which yields the wide distribution of δ. The damping factor also depends on temperature, which is maximum around T_f and on the other hand I^* decreases monotonically.

Our view of $Fe_{0.7}Al_{0.3}$ is such a heterogeneous magnetic state that the ferromagnetic clusters in the alloy are weakly linked with each other above T_f in the ferromagnetic phase. Since the clusters are infinitely connected, we can define as the normal ferromagnetic state above T_c^{inv}. T_c^{inv} is defined as the temperature below which the bulk magnetization starts to decrease with decreasing temperature. When temperature goes across T_c^{inv}, the clusters are separated from each other by the break of the weakly linked bonds and the magnetic phase becomes the mixture of the large clusters and the spin glass part of the rest. This picture is consistent with the fact of thermal evolution of both I^* and σ_I. When temperature approaches T_f, where σ_I is maximum, the δ is too small to maintain a finite mesoscopic field keeping the bulk field to the unique direction. Eventually no depolarization occurs in the ZFC process due to the fact that neutron polarization follows adiabatically in the sample. In the FC process, the cluster size of δ is smaller and furthermore the spin glass like region coexists, judging from the small I^* as well as the large σ_I values, although the average magnetization in each domain directs towards the external field.

Thus the present depolarization experiments have shown for the first time the existence of mesoscopically heterogeneous magnetic state in the RSG phase below T_f, which should be emphasized to occur only in the strongly frustrated RSG materials. It is also emphasized here that it differs from so called the mixed state of the RSG in view of the mean field theory.

The depolarization result of an amorphous $Fe_{0.7}Mn_{0.3}$ RSG resembles that of the $Fe_{0.7}Al_{0.3}$ glass, in the sense that the depolarization decreases below T_f, or I^* substantially decreases which was given by the analysis of the data of the gaussian λ

dependence. Although the $Ni_{1-x}Mn_x$ data are not complete, a similar thermal evolution was observed.

§ 4. Conclusions

Spin glass is an intrinsic effect of the topological spin disorder superposed by the competition of magnetic interactions, which now becomes a well established physical concept. On the other hand, the reentrant spin glass behavior is yet a subtle problem, which brings controversies in theories and experiments to investigate the nature of the phase transition, the magnetic ground state and so forth. The depolarization method measuring the mesoscopic magnetic induction clearly indicates the existence of the phase transition from the ferromagnetic LRO to the spin glass like phase. The depolarization experiments also show the evidence that the magnetic state of the RSG phase differs from materials to materials reflecting the degree of the frustration, even though any difference is difficult to be seen in thermal evolution of these bulk magnetizations.

In the weakly frustrated systems of the $Au_{1-x}Fe_x$ alloy, the RSG phase is realized by freezing randomly oriented transverse spins keeping the longitudinal component ordered. Thus the total magnetization in each domain is canted with each other, which creates an extra anisotropy preventing the free motion of the domain walls. The mesoscopic field substantially decreases as the temperature decreases across T_f into the RSG phase. Furthermore the depolarization curves of the exponential function of λ are well interpreted by the depolarization data from the ferromagnetic state with many small domains. It is therefore concluded that the magnetic state of the RSG phase of the weakly frustated system is determined to be the mixed state just as the mean field theory predicted.

On the other hand, in the strongly frustrated system of $Fe_{1-x}Al_x$ alloy, the RSG phase is the heterogeneous state consisting of small ferromagnetic domains floating in the spin glass like media. This magnetic state already develops in the ferromagnetic phase far above T_f. The magnetization decreases with decreasing temperature across T_c^{inv}, and correspondingly the mesoscopic field detected by the depolarization also decreases associated with the large increase of damping factor σ_I. Since a heuristic relation of σ_I and I^* in the ferromagnet does not hold in this case, the drastic increase of σ_I directly corresponds to the large δ distribution even in the ferromagnetic state. This experimental fact gives a conclusion that infinitely connected ferromagnetic domains are broken to be dispersed in the spin glass like medium, and eventually no mesoscopic steady field exists below T_f.

This physical picture of the onset of the reentrance from the long range ordered to the strongly frustrated spin glass phase is consistent with the concept of the local mean field theory studied by Saslow and Parker.[19] The magnetic defects which strongly frustrate the host LRO, lead to the weakning of the ordered magnetism to break up the infinite connection of the domains. On the other hand, in the weakly frustrated RSG, many defects are required to help the reentrance, but eventually the ferromagnetic matrix becomes noncollinear spin arrangement in the RSG phase, which is equivalent to the mean field picture of the magnetic state of the mixture of

the randomly oriented frozen transverse and the long range ordered longitudinal components.

Finally the time dependent depolarization experiment was carried out to see how dispersed domains grow by thermal activation of the domain wall motion by changing temperature, incubating time in the applied field and the applied field value itself. Either a fully polarized curve in the strongly frustrated RSG or a monotonic λ dependent curve of the exponential function in the weakly frustrated RSG for the ZFC changes to the damped oscillatory curve of the λ dependence when the external field exceeds threshold field H_c. H_c substantially decreases with the increase of temperature approaching T_f. This experiment indicates that the growth of ferromagnetic domains starts to develop above H_c and these domains are stabilized even when the field is removed. The domain growth rate is also time dependent and eventually it goes to the saturated values with the rate approximated by the function of $A-Bt^{-\alpha}$. The study of the kinetics of growth of the order in the spin glass is not an easy task. At present no reliable theory exists on this specific subject. However such an experiment itself is attractive because precise controls of various external parameters are absolutely necessary in order to obtain any conclusive result. Therefore this subject becomes more fascinating problem for us to challenge in future.

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References

1) K. Binder and A. P. Young, Rev. Mod. Phys. **58** (1986), 801.
2) S. F. Edwards and P. W. Anderson, J. of Phys. **F5** (1975), 965.
3) M. Gabay and G. Toulouse, Phys. Rev. Lett. **47** (1981), 201.
4) J. R. L. De Almeida and D. J. Thouless, J. of Phys. **A11** (1978), 983.
5) D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. **35** (1975), 1792.
6) H. Malletta, *Lecture Notes in Physics* **192** (Springer, Berlin, 1983), p. 90.
7) G. Aeppli, S. M. Shapiro, R. J. Birgeneau and H. S. Chen, Phys. Rev. **B28** (1983), 5160.
8) Y. Ishikawa, Y. Endoh and K. Inoue, *Neutron Scattering in Nineties* (IAEA Vienna, 1985), p. 282.
9) O. Halpern and Y. Holstein, Phys. Rev. **59** (1941), 960.
10) M. Th. Rekveldt, Z. Phys. **256** (1973), 391.
11) S. Mitsuda and Y. Endoh, J. Phys. Soc. Jpn. **54** (1985), 1570.
12) F. Mezei, *Lecture Notes in Physics* **128** (Springer, Berlin, 1979), p. 1.
13) Y. Endoh, Y. Sasaki, H. Ono, S. Mitsuda and H. Fujimoto, Nucl. Instr. & Meth. **A240** (1985), 115.
14) G. Parisi, Phys. Rev. Lett. **43** (1979), 1754.
15) S. Mitsuda, H. Yoshizawa and Y. Endoh, submitted to Phys. Rev. B

16) S. Mitsuda, private communications.
17) I. Mirebeau, S. Itoh, S. Mitsuda, T. Watanabe, Y. Endoh, M. Hennion and R. Papoular, Phys. Rev. **B41** (1990), 11405.
18) I. Mirebeau, S. Itoh, S. Mitsuda, T. Watanabe, Y. Endoh, M. Hennion and P. Calmettes, J. Appl. Phys. **67** (1990), 5232.
19) W. M. Saslow and G. N. Parker, Phys. Rev. Lett. **56** (1986), 1074.