Linear and Nonlinear Susceptibilities of Amorphous Fe-RE-Zr (RE= Ho, Dy, Er) Alloys

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Linear and nonlinear AC susceptibilities are examined for mechanically alloyed amorphous $(\text{Fe}_{1-x}\text{RE}_x)_{0.7}\text{Zr}_{0.3}$ ($x \leq 0.4$; RE=Ho, Dy and Er) which exhibit spin freezing behaviors due to the random magnetic anisotropy of RE atoms. A negative divergent behavior of nonlinear susceptibility χ_2 is observed for the specimens with $x=0.1$, similar to a case of a spin-glass ordering. On the other hand, for the specimens with $x=0.3$, χ_2 is found to show an antisymmetric anomaly, indicating a ferromagnetic ordering. Such behaviors reveal a reentrant-spin-glass-like ordering character of the specimens with $x=0.3$ at zero DC field. The magnetic phase diagrams are proposed and the ordering characteristics of the systems are discussed with a systematic change of the RE concentration.

Key words: random magnetic anisotropy, correlated spin-glass, amorphous alloy, mechanical alloying, nonlinear susceptibility, ac susceptibility.

1. Introduction

In amorphous transition metal (TM) - heavy rare earth (RE) alloys, the random magnetic anisotropy (RMA), which is possessed by non-S state RE element sitting in the asymmetric electrostatic environment due to the random atomic arrangement in amorphous alloys, suppresses the ferrimagnetic spin structure and invokes spin-glass-like behaviors. Some characteristic noncollinear spin structures and spin freezing behaviors could be induced by RMA, such as correlated spin glass (CSG) state proposed by Chudnovsky et.al. through the theoretical investigation for a continuous-symmetry ferromagnet with RMA [1]. The ordering of these materials is an attractive subject from the view point of cooperative dynamics, but may also be an important basic property for the application to magneto optical recording media.

Meanwhile, our recent measurements of magnetization and AC susceptibility under DC magnetic fields for amorphous Fe-RE-Zr alloys suggested the existence of some characteristic ordered state which originates from RMA [2-3]. In the present work, linear and nonlinear AC susceptibilities are examined in order to do the precise investigation for the magnetic ordering of $(\text{Fe}_{1-x}\text{RE}_x)_{0.7}\text{Zr}_{0.3}$ ($x \leq 0.4$; RE=Ho, Dy and Er) amorphous alloys. Originally, nonlinear susceptibility χ_2 was introduced theoretically as a reliable marker of the phase transition into a spin-glass state by the negative divergence at the critical point [4-5]. Experimentally, the measurement of χ_2 has been successfully applied for some complex magnets [6-7], showing the validity of χ_2 analysis for the ordering of such systems. Thus, χ_2 is a useful indicator of the

magnetic ordering in the present systems.

2. Experimental

Amorphous Fe-RE-Zr samples were prepared by mechanical alloying in a planetary ball mill under an Ar gas atmosphere. AC magnetic measurements were carried out by a mutual inductance method. Nonlinear susceptibility χ_2 , which is the third order expansion coefficient of the magnetization to the external field, was experimentally examined by nonlinear magnetic response $M'_{3\omega}$ ($\sim -1/4 \chi_2 h^3$), the third harmonic in-phase component of magnetization to an AC field $h \sin \omega t$. χ_2 anomaly at thermal equilibrium condition should be examined by the static limit of $M'_{3\omega}-T$ curve.

3. Results and Discussion

First, the $H-T$ phase diagrams for amorphous $(\text{Fe}_{1-x}\text{Ho}_x)_{0.7}\text{Zr}_{0.3}$ with $x=0.1, 0.3$ and 0.4 were examined through AC susceptibility measurements parallel and perpendicular to DC magnetic field, in the same way with that for $(\text{Fe}_{1-x}\text{Dy}_x)_{0.7}\text{Zr}_{0.3}$ previously examined [3]. The proposed phase diagrams are summarized in Figs. 1(a)–1(c).

For $x=0.1$, the phase diagram is given in Fig. 1(a). At low temperatures, correlated spin glass (CSG) phase, which is characterized by spin freezing with a large ferromagnetic correlation length but zero net magnetization [1], would appear in the magnetic field, showing two-step spin freezing of transverse and longitudinal components, such like G-T and A-T lines in a vector spin glass system. By increasing magnetic field or temperature, the CSG phase would transform to a ferromagnetic state with wandering axes (FWA), which is predicted to occur for a random anisotropic magnet [1]. The phase diagram is qualitatively similar to that of the amorphous Fe-Dy-Zr alloy with $x=0.1$ [3].

Under zero magnetic field, the ordering of amorphous $(\text{Fe}_{1-x}\text{Ho}_x)_{0.7}\text{Zr}_{0.3}$ with $x=0.1$ would proceed in a single step from PM to CSG, as seen from Fig. 1(a). In order to confirm the ordering at zero DC field, the temperature dependence of AC susceptibilities χ^* ($=\chi' - i\chi''$) and nonlinear magnetic response $M'_{3\omega}$ were examined at zero DC field. The results are given in Fig. 2. The $\chi''-T$ curves exhibit a maximum at a temperature T_p , which was considered to be the freezing temperature [3], but shows no remarkable frequency dependence within the present frequency range. χ'' also shows a maximum around T_p , indicating an increase of magnetic viscosity around T_p . On the other hand, the $M'_{3\omega}-T$ curve at $f=311$ Hz exhibits a sharp positive peak and the peak height

Fig. 1 Phase diagrams for the specimens with (a) $x=0.1$, (b) $x=0.3$ and (c) $x=0.4$. Open and closed symbols were obtained from the data of transverse and longitudinal AC susceptibilities, respectively.

Fig. 2 Temperature versus AC susceptibility χ^* and nonlinear magnetic response $M'_{3\omega}$ for $(\text{Fe}_{0.9}\text{Ho}_{0.1})_{0.7}\text{Zr}_{0.3}$.

becomes larger at a lower frequency $f=37$ Hz, as shown in the figure. Such behaviors suggest a negative divergence of χ_2 at the thermal equilibrium condition, which is a useful indicator of spin-glass ordering [4-5]. The divergent-like behavior of the $M'_{3\omega}-T$ curve is thus

considered to correspond to the critical phenomenon of the transition into the CSG phase, although singularity of χ_2 for the CSG ordering is unknown theoretically. A discrepancy between the peak temperatures of χ' and $M'_{3\omega}$ could be attributed to the non-zero measuring frequency of $M'_{3\omega}$.

For $x=0.3$, the phase diagram is shown in Fig. 1(b) and is qualitatively similar to that for Fe-Dy-Zr system with $x=0.3$ [3]. In the phase diagram, an intermediate phase can be seen even at zero magnetic field, different from the case of the specimens with $x=0.1$. In fact, there are two subnetworks of Fe and RE atoms coupled antiferromagnetically each other in the systems. The subnetwork magnetization of RE atoms (Fe atoms) are dominant in the specimen with $x=0.3$ ($x=0.1$), while they are almost cancelled out around $x=0.2$ [3]. The intermediate phase is considered to be FWA caused by the unbalanced moments of RE atoms which could be coupled ferromagnetically through the antiferromagnetic interaction with the Fe atoms. The origin of successive ordering character of the FWA phase under DC field was discussed previously [3].

Under zero magnetic field, the ordering of the specimen with $x=0.3$ is considered to proceed in two steps from PM to FWA and from FWA to CSG, as seen in Fig. 1(b). Such ordering character is similar to that of a reentrant-spin-glass. Shown in Fig. 3 are AC susceptibility and nonlinear magnetic response performed for the specimen with $x=0.3$ at zero DC field. In the figure, AC susceptibility χ' shows a rapid increase as decreasing temperature from the PM state and exhibits a peak around 95 K at $f=311$ Hz, suggesting a phase transition into the FWA phase. At a lower frequency $f=37$ Hz, the peak shifts to a lower temperature side with a little increase in the magnitude below the peak temperature T_p where χ'' exhibits a maximum, indicating a phase transition accompanied with magnetic viscosity. χ'' , in addition, shows a shoulder around 50 K, where χ'' also begins to increase again. These anomalies of χ^* may be related to the CSG ordering at T_{csg} which is considered to be about 60 K as seen from Fig. 1(b). On the other hand, the nonlinear magnetic response $M'_{3\omega}$ exhibits a rapid increase with positive sign by decreasing temperature and abruptly changes the sign from positive to negative around T_p at $f=108$ Hz. The shape of anomaly is unchanged with decreasing frequency down to 11 Hz. Such an antisymmetric anomaly is qualitatively in good agreement with χ_2 anomaly for the ferromagnetic ordering predicted theoretically [5] and quite different from that observed for the specimen with $x=0.1$. The phase transition at T_p under zero field is therefore confirmed to be into the FWA phase, suggesting the reentrant-spin-glass-like ordering character. In this way, the qualitative difference of ordering characteristics between the specimens with $x=0.1$ and 0.3 is successfully revealed by $M'_{3\omega}$ rather than χ^* . $M'_{3\omega}$, however, shows no remarkable anomaly around T_{csg} . Although χ_2 has been experimentally examined for some ferromagnetic reentrant spin glass systems [8], the anomaly at the reentrant-spin-glass transition point is usually very small and non-divergent, while that for the ferromagnetic phase transition is relatively

Fig. 3 Temperature versus AC susceptibility χ^* and nonlinear magnetic response $M'_{3\omega}$ for $(\text{Fe}_{0.7}\text{Ho}_{0.3})_{0.7}\text{Zr}_{0.3}$.

Fig. 4 Temperature versus AC susceptibility χ^* and nonlinear magnetic response $M'_{3\omega}$ for $(\text{Fe}_{0.6}\text{Ho}_{0.4})_{0.7}\text{Zr}_{0.3}$.

large and divergent. It is difficult to observe such a small anomaly at the present sensitivity range.

Next, H - T phase diagram for $x=0.4$ is seen in Fig. 1(c), although the specimens with $x>0.3$ was not examined in our previous work [3]. As seen from the figure, no ferromagnetic phase in the magnetic field is found through the measurement, different from the specimens with $x=0.1$ and 0.3 . However, the freezing of transverse and longitudinal spin components to the magnetic field seems to occur in two steps, such as in a vector spin glass system. In order to clarify the ordering, χ^* and $M'_{3\omega}$ are measured at zero DC field and the results are shown in Fig. 4. In the figure, χ' shows a maximum at a temperature T_p , which becomes lower with decreasing frequency ω down to 37 Hz. On the other hand, χ'' rapidly increases near $T_p(\omega)$ and appears to make an inflection at $T_p(\omega)$, indicating a rapid increase of the magnetic loss below $T_p(\omega)$. Such behaviors of χ^* indicate a spin freezing at $T_p(\omega)$ within the time scale of observation. On the other hand, the $M'_{3\omega}$ - T curve at $f=311$ Hz shows a positive peak near T_p and changes the sign as temperature decreases. The $M'_{3\omega}$ - T curve slightly increases the peak amplitude at a lower frequency of $f=108$ Hz. Such behaviors of $M'_{3\omega}$ let us suspect a negative divergence of χ_2 but the further analyses at lower frequency ranges are

essential in order to estimate the static limit. Although the anomaly of χ_2 is not well clarified at the present stage, the ordering characteristics of the specimen with $x=0.4$ is considered to be of a conventional spin glass, taking the behaviors of χ^*-T into account.

CSG phase is predicted to be realized in a ferromagnet with weak RMA [1]. The CSG phase seen for $x=0.1$ thus originates from the weak RMA due to the lower Ho concentration in the Fe-based ferromagnet. On the other hand, for $x=0.4$, CSG phase no longer appears. Such a feature is also confirmed for the specimens with RE=Dy and Er. This is because the strong RMA is caused by the relatively higher RE concentration. Strong RMA induces conventional spin-glass-like nature instead of CSG phase [9]. The RKKY interactions between RE atoms are considered to be another candidate for the origin of the spin-glass-like ordering. The transition into the CSG phase, however, occurs for $x=0.3$. Both effects of RMA and RKKY interactions, considering the less RE concentration, must be weaker than that for $x=0.4$, so that the CSG phase could be realized. The CSG ordering for $x=0.3$ is reentrant-type. Such a double transition has been also found in some amorphous RE-TM alloys [10] but the mechanism is not yet understood. The reentrant characteristic of the CSG ordering should be noted the resemblance between a random anisotropic magnet and spin glass, although the origin is quite different each other.

4. Summary

A negative divergent behavior and an antisymmetric anomaly of χ_2 are found for amorphous $(\text{Fe}_{1-x}\text{Ho}_x)_{0.7}\text{Zr}_{0.3}$ with $x=0.1$ and 0.3 , respectively. It is successfully distinguished whether each ordering character is spin-glass-like or reentrant-spin-glass-like at zero DC field. The specimen with $x=0.4$ is found to be a conventional spin-glass rather than CSG. Also for the other Fe-RE-Zr systems with RE=Dy and Er, the ordering characteristics are confirmed to be qualitatively similar to those of the Fe-Ho-Zr system.

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