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Ageing and memory effects in the weak random anisotropy magnets amorphous NdGdFe and HoGdFe

Toshiaki Saito[1,2], **Ai Emura**[1] **and Koji Hanashima**[1]

[1] Department of Physics, Faculty of Science, Toho University, Funabashi City, Chiba 274-8510, Japan
[2] Research Center for Materials with Integrated Properties, Toho University, Funabashi City, Chiba 274-8510, Japan

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Abstract
We experimentally examined the ageing phenomena in typical weak random anisotropy magnets (weak RAMs), amorphous NdGdFe and HoGdFe, with a small ratio of the random anisotropy (D) to the ferromagnetic exchange (J) ($D/J \ll 1$). These weak RAMs have very long average relaxation time, two or three orders longer than that of spin glasses (SGs) around the transition temperature, and also have a very large ac excitation field (h_0) dependence of the ac susceptibility. Measuring the imaginary part of the ac susceptibility at frequency of 0.5 Hz and h_0 of 0.3 Oe by using two temperature-change protocols, we observed the memory and rejuvenation effects as reported in SGs, but the effects are weaker in the present weak RAMs, suggesting that the picture of the hierarchical structure of the free energy space is also effective in weak RAMs as in SGs, but it may have smaller barrier heights than those of SGs.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The magnetic random anisotropy system has frustration due to competition between the uniaxial random anisotropy (D) and the ferromagnetic exchange (J). Weak random anisotropy magnets (weak RAMs) with small D/J ($\ll 1$) have several characteristic properties [1–4] different from both spin glasses (SGs) and 'strong RAMs' [5] with larger D/J (>1). In particular, the dynamical behaviour around the transition temperature (T_c) is remarkable [3]; the magnitude of the imaginary part (Im χ, 90° out of phase component) of the ac susceptibility in weak RAMs such as amorphous (a-)NdGdFe and a-HoGdFe is comparable to that of the real part (Re χ) or even larger than Re χ, which indicates that the average relaxation time is two or three orders larger than that of SGs. Recently [6], we measured the ac field dependence of the ac susceptibility for the weak RAM, a-$(\mathrm{Nd}_{0.3}\mathrm{Gd}_{0.7})_{19}\mathrm{Fe}_{81}$ (T_c is about 290 K), to investigate the origin of the longer relaxation in weak RAMs. The magnitude of ac susceptibility strongly depends on the excitation field, h_0; with increasing h_0, both Re χ and Im χ become several times larger, and the ratio Im χ to Re χ increases for $h_0 < 1$–2 Oe, causing the longer relaxation times in weak RAMs. In addition, temperature width of the peak of Re χ broadens extremely and the peak temperature of Im χ shifts continuously to the lower-temperature side with increasing h_0.

This large h_0 dependence may arise from the free energy structure with small height of barriers due to the small D/J in weak RAMs with weak frustration, compared with that of SGs or strong RAMs; such small barriers with energy height of order of h_0 ($\sim$1 Oe) in weak RAMs may be rather easily surmounted and the system can move to some other local minimum even located far away, which may cause the long relaxation time. Assuming the hierarchical structure of the free energy space also in weak RAM as in SG, we may also explain the broadening of temperature width of Re χ [6].

In this paper, in order to investigate experimentally the structure of free energy in weak RAMs, we examined the ageing phenomena in weak RAMs compared to those of SGs. Although the ageing phenomena (memory and rejuvenation effects, etc) have been extensively studied in SGs [7, 8] and in many glassy systems [9] by using many temperature-change protocols, the ageing phenomena in weak RAMs have not been reported as far as we know.

2. Experimental details

This films of a-$(\mathrm{Nd}_{0.9}\mathrm{Gd}_{0.1})_{18}\mathrm{Fe}_{82}$ and a-$(\mathrm{Ho}_{0.6}\mathrm{Gd}_{0.4})_{21}\mathrm{Fe}_{79}$ (the thicknesses were $\sim$1.5 μm) were prepared by using a magnetron sputtering technique as in our previous works [2–4]. The coherent (not random but uniform) anisotropy of the sample was enough small not to induce the ferromagnetic-like (FWA) phase [1, 4] at static magnetic field $H = 0$ in these samples. Ageing phenomena were observed by measuring the ac susceptibility, in particular, Im χ, by using a SQUID system. The ac excitation field has a frequency (f) of 0.5 Hz and the amplitude (h_0) was 0.3 Oe (small magnitude which hardly affects χ), which was applied parallel to the film plane. The protocols of temperature change for observation of ageing were (1) continuous temperature change with an intermittent stop at T_{ageing} below T_{c}, and (2) negative and positive temperature cycle at T_{ageing} as described below.

3. Results and discussion

First, we measured the ac susceptibility for a-$(\mathrm{Nd}_{0.9}\mathrm{Gd}_{0.1})_{18}\mathrm{Fe}_{82}$ on cooling the sample from 300 K above T_{c} ($\sim$221 K) down to 70 K at a constant cooling rate of 1 K min^{-1}, and then heating it back continuously at the same rate. The two curves of χ obtained are almost the same. In figure 1, the 'reference curve' of Im χ is shown. We then repeated the measuring, but now stopped at an intermediate temperature T_{ageing} (=210 K) for a certain time, $t_{w0} = 950$ min, only during cooling. During this time t_{w0}, both Re χ and Im χ relax downwards due to ageing. In figure 1, only Im χ is represented because the relative amount of variation of Im χ is much larger than Re χ. In the following we concentrate only on Im χ. After the ageing at 210 K, the cooling procedure resumed. We notice that Im χ merges back with the reference curve at $\sim$200 K (only 10 K below T_{ageing}), indicating that ageing at 210 K does not influence the result at lower temperatures; the chaos effect, as reported in the Heisenberg SG sample $\mathrm{CdCr}_{1.7}\mathrm{In}_{0.3}\mathrm{S}_4$ [7], is also observed in this weak RAM.

When the system is reheated at the same rate, the magnitude of Im χ decreases from the reference curve only around T_{ageing} (from 200 to 220 K), which suggests that the 'memory' of ageing at 210 K has not been erased by the cooling procedure, though the memory is not perfect, as observed in SGs [7], in which even the dip in Im χ at T_{ageing} is exactly recovered.

Second, we examined the negative temperature-cycle protocol for the same sample, a-$(\mathrm{Nd}_{0.9}\mathrm{Gd}_{0.1})_{18}\mathrm{Fe}_{82}$. This protocol consists of the following three stages; (1) the system is

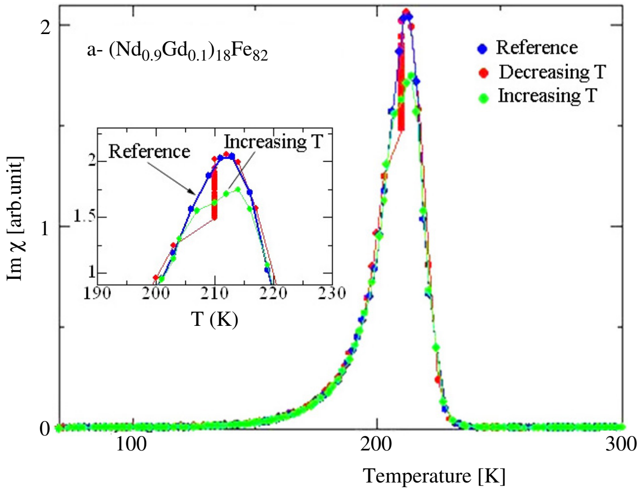

Figure 1. Imaginary part of the ac susceptibility (Im χ) for a weak random anisotropy magnet, a-$(\mathrm{Nd}_{0.9}\mathrm{Gd}_{0.1})_{18}\mathrm{Fe}_{82}$. The measuring frequency is 0.5 Hz, and the ac field is 0.3 Oe. The reference curve was measured continuously at a constant cooling and heating rate of 1 K min^{-1} from 300 K. The measurements of Im χ at the same cooling (decreasing T) and heating (increasing T) rate were performed, except for ageing at 210 K for 950 min (only when decreasing T).

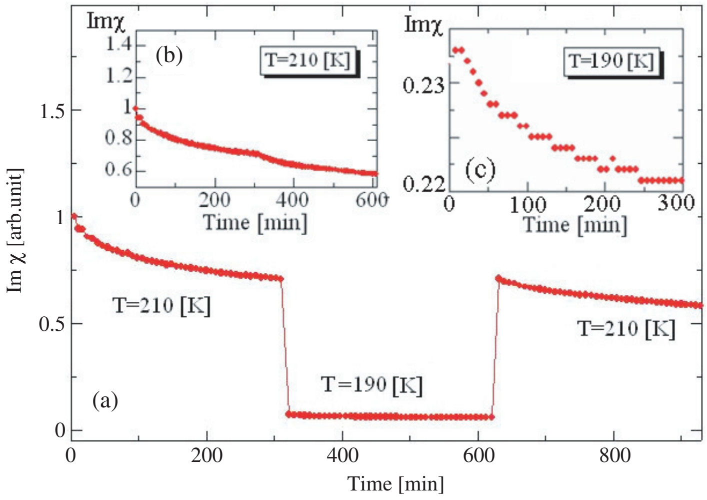

Figure 2. (a) Effect of a negative temperature cycling ($\Delta T/T_{\mathrm{ageing}} \sim 0.1$) on the time dependence of Im χ ($f = 0.5$ Hz, $h_0 = 0.3$ Oe) for a-$(\mathrm{Nd}_{0.9}\mathrm{Gd}_{0.1})_{18}\mathrm{Fe}_{82}$ ($T_c \sim 221$ K). The inset (b) shows a plot of the data points during the first period and the third period at 210 K by removing the second period data at 190 K and by shifting the timescale of the third period. The inset (c) shows the data points during the second period on an extended Im χ scale.

cooled down from 300 K (above T_c) to $T_{\mathrm{ageing}} = 210$ K at 10 K min^{-1}, and is kept at 210 K for a time $t_{w1} = 300$ min, (2) the temperature is temporarily reduced to 190 K ($\Delta T/T_{\mathrm{ageing}} \sim 0.1$) for a time $t_{w2} = 300$ min, (3) then it is set back to 210 K (for 300 min).

Figure 2(a) presents the results on Im χ. (We note that the ac field was zero during the initial cooling process from 300 K to $T_{\mathrm{h}} = 215$ K (5 K above T_{ageing}) and $h_0 = 0.3$ Oe from T_{h} to T_{ageing}. The following result was the same under the condition of $T_{\mathrm{h}} > T_{\mathrm{ageing}}$.) During

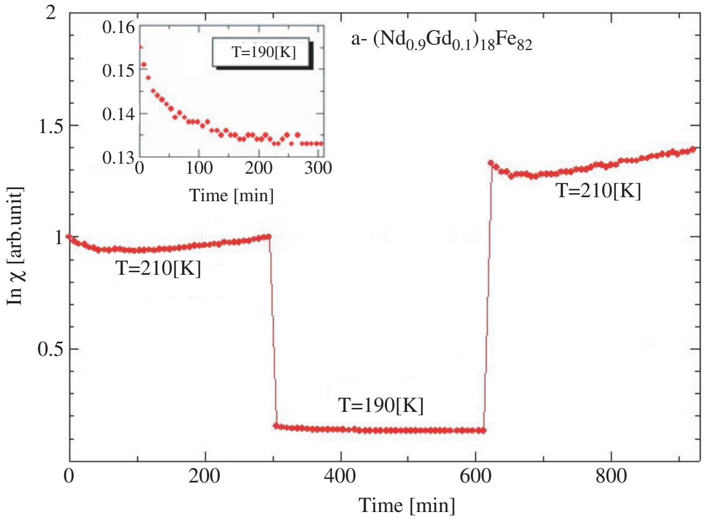

Figure 3. Im χ as a function of time ($f = 0.5$ Hz, $h_0 = 0.3$ Oe) for a-$(Nd_{0.9}Gd_{0.1})_{18}Fe_{82}$ ($T_c \sim 221$ K). It is noted that the excitation field was initially applied at $t = 0$ (h_0 was 0 during the initial cooling process from 300 to 210 K). The inset shows the data points during the second period on an extended Im χ scale.

the first period t_{w1}, the standard relaxation characteristic of ageing is observed. At the first temperature change, a new decay of Im χ occurs at 190 K, as shown in the inset (c) of figure 2 (the second stage), indicating that new ageing starts when the temperature is lowered. At the beginning of the third stage, Im χ returns almost immediately to the standard relaxation with a weak rejuvenation jump. The negative temperature cycle during t_{w2} has not erased the ageing of the first stage during t_{w1}, and then the relaxation is resumed when the temperature is raised back to 210 K (the memory effect) as shown in the inset (b) of figure 2. In the inset, data points at the second stage during t_{w2} at 190 K are removed, and Im χ at the first stage and Im χ at the third stage are directly connected by shifting the timescale of the third period. The best continuation of the data groups of the different two stages seems to require the addition of a small extra time t_{eff} ($\ll t_{w2}$) between the two data groups, even if the small jump of Im χ at the beginning of the third period is excluded (weak memory effect).

These ageing phenomena observed in a weak RAM system imply that the hierarchical structure of the free energy space and a continuous ramification of the metastable states with decreasing temperature are also present in a weak RAM as in SGs. Similar weak rejuvenation by raising the temperature and weak memory effect as shown in figure 2 have been reported for small $\Delta T/T_{\mathrm{ageing}}$ ($\sim$0.025) in an SG ($CdCr_{1.7}In_{0.3}S_4$) [8]. Since the ageing process at higher states has also occurred if the decreasing temperature width is small enough (if $\Delta T/T_{\mathrm{ageing}}$ is small enough), the system will have effectively aged for t_{eff}. In the present weak RAM, the weak rejuvenation and the weak memory effect are observed even for $\Delta T/T_{\mathrm{ageing}} \sim 1.0$, which temperature width is not small, at least for an SG ($CdCr_{1.7}In_{0.3}S_4$) [8]. However, the probability of processes across barriers existing at higher temperature may increase if the effective barrier heights are averagely smaller in this weak RAM. This hierarchical picture of free energy space with smaller barrier heights is consistent with our previous experiment [6] in which very strong h_0 dependence of ac susceptibility is observed in weak RAMs as introduced in section 1.

Figure 3 shows the result on Im χ of the same sample in the same condition as in figure 2 except that the ac field was zero during the first cooling process from 300 K ($>T_c$) to 210 K

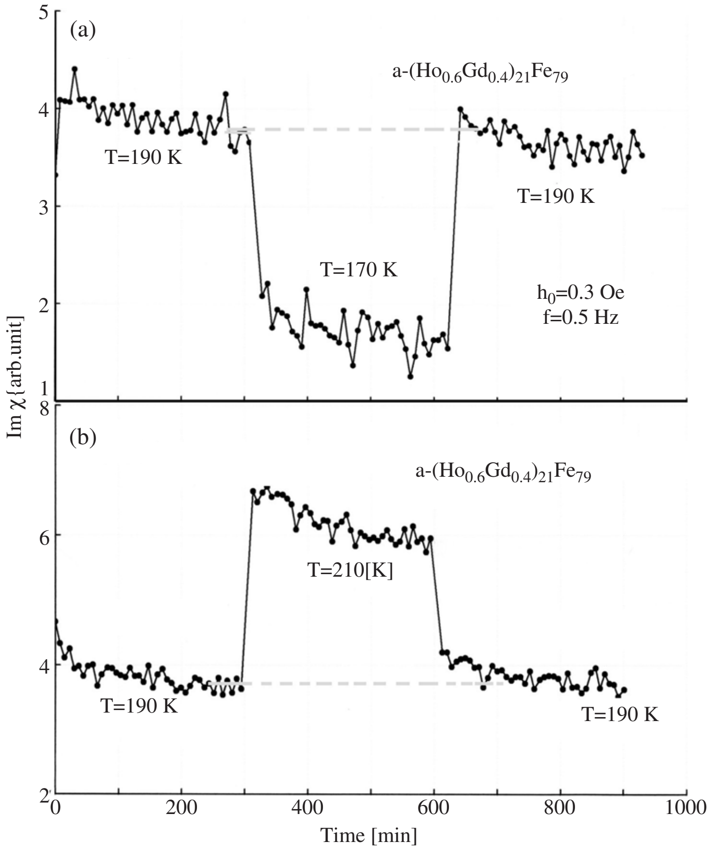

Figure 4. Effect of (a) negative temperature cycling and (b) positive temperature cycling ($\Delta T/T_{\mathrm{ageing}} \sim 0.1$) on the time dependence of Im χ ($f = 0.5$ Hz, $h_0 = 0.3$ Oe) at $T_{\mathrm{ageing}} = 190$ K for a-$(\mathrm{Ho}_{0.6}\mathrm{Gd}_{0.4})_{21}\mathrm{Fe}_{79}$ ($T_{\mathrm{c}} \sim 250$ K).

($T_{\mathrm{h}} = T_{\mathrm{ageing}}$). We found that Im χ does not decrease but continues to increase during the first period t_{w1}, though Im χ initially relaxes at $t \sim 0$. The surprise is that, when the temperature is lowered to 190 K at the beginning of the second stage, the new normal ageing occurs during t_{w2} (see the inset of figure 3) independently of the process in the first stage, and when the temperature is raised back to 210 K at the beginning of the third stage, the increasing of Im χ resumes and Im χ continues to increase during t_{w3} except for the rejuvenation jump at the beginning of the stage. Such distinct temperature independence has not been reported in SGs and other glassy systems as far as we know. The interpretation of this result at 210 K seems to be difficult, but we guess that a kind of 'supercooling' occurs during the initial cooling from 300 K above T_{c} (=221 K) to 210 K without ac excitation field, and then the system returns to the inherent state at 210 K after the ac field is applied. To clarify this phenomenon, however, further experiments and discussion are needed.

In figure 4 is shown Im χ measured for another weak RAM, a-$(\mathrm{Ho}_{0.6}\mathrm{Gd}_{0.4})_{21}\mathrm{Fe}_{79}$ ($T_{\mathrm{c}} \sim$ 250 K) with slightly smaller D/J than that of a-$(\mathrm{Nd}_{0.9}\mathrm{Gd}_{0.1})_{18}\mathrm{Fe}_{82}$ due to the large amount of Gd ($D \sim 0$). (The signal-to-noise ratio of a-HoGdFe in figure 4 is smaller than that of a-NdGdFe in figure 2 because the total magnetization of a-HoGdFe is rather small.) Similar ageing effects to a-NdGdFe, which seem to be somewhat weaker due to smaller D/J, are again observed in this weak RAM. For the positive T-cycling protocol (figure 4(b)), the memory of ageing for t_{w1} at the first stage is almost erased when temperature is raised from 190 to 210 K as in an SG [8]. To study the D/J dependence definitely on the ageing effect in weak RAMs, more systematic investigation is needed by controlling D/J, for instance, by observing the ageing effects of a series of a-$(\mathrm{Nd}_{1-x}\mathrm{Gd}_x)_{18}\mathrm{Fe}_{82}$ or a-$(\mathrm{Ho}_{1-x}\mathrm{Gd}_x)_{21}\mathrm{Fe}_{79}$ containing different Gd content (x) (D/J becomes smaller with increasing x).

In conclusion, we observed similar but slightly weaker ageing phenomena in weak RAMs to those of SGs, which suggests that the picture of hierarchical structure of the free energy space is also effective in weak RAM as in SGs, but it may have smaller barrier heights than those of SGs.

References

[1] Chudnovsky E M and Serota R A 1982 *Phys. Rev.* B **26** 2697

[2] Saito T, Matsumaru Y, Shinagawa K and Tsushima T 1994 *J. Magn. Magn. Mater.* **130** 347

[3] Saito T, Suto A and Takenaka S 2004 *J. Magn. Magn. Mater.* **272–276** 1319

[4] Saito T, Mochiduki K, Masago E, Shinagawa K and Tsushima T 1999 *J. Appl. Phys.* **85** 4509

[5] Dieny B and Barbara B 1986 *Phys. Rev. Lett.* **57** 1169
Sellmyer D J and Nafis S 1986 *Phys. Rev. Lett.* **57** 1173

[6] Saito T and Hanashima K 2006 Large ac field dependence of dynamic susceptibility in a weak random anisotropy magnet a-NdGdFe *Proc. ICM2006* at press

[7] Jonason K, Vincent E, Hammann J, Bouchaud J P and Nordblad P 1998 *Phys. Rev. Lett.* **81** 3243

[8] Vincent E, Bouchaud J P, Hammann J and Lefloch F 1995 *Phil. Mag.* B **71** 489

[9] For example, Bellon L, Ciliberto S and Laroche C 2000 *Europhys. Lett.* **51** 551