Anomaly in out-of-phase component of ac susceptibility of the cluster-glass system $Fe_xZn_{1-x}F_2$ with x = 0.26 and 0.10

1998 J. Phys.: Condens. Matter 10 L711

(http://iopscience.iop.org/0953-8984/10/43/001)

J. Phys.: Condens. Matter **10** (1998) L711–L716. Printed in the UK PII: S0953-8984(98)97228-9

LETTER TO THE EDITOR

Anomaly in out-of-phase component of ac susceptibility of the cluster-glass system $Fe_xZn_{1-x}F_2$ with $x = 0.26$ and 0.10

J Satooka† and A Ito
Graduate School of Humanities and Sciences, Ochanomizu University, Bunkyo-ku, Tokyo 112-8610, Japan

Received 2 September 1998

Abstract. The cluster-glass (CG) system $Fe_xZn_{1-x}F_2$ with $x = 0.26$ and 0.10 has been studied by ac susceptibility measurements in bias dc fields, $H_{dc} = 0 \sim 10$ kG. The temperature dependence of the out-of-phase component of the ac susceptibility χ'' of both samples has a strange structure with some irregularities in the temperature range $T < 8$ K. The temperatures where the irregularity appears do not depend on either H_{dc} or Fe concentration x, but do depend strongly on frequency. This behaviour of χ'' of the CG system $Fe_xZn_{1-x}F_2$ with $x = 0.26$ and 0.10 in a low temperature region is interpreted to originate from the contribution of small clusters fluctuating dynamically.

Spin-glass (SG) freezing has been considered to be a phenomenon which originates from random exchange frustration [1]. Some examples of SG freezing have been reported in dilute systems, such as $Eu_xSr_{1-x}S$ [2] and $Mn_xMg_{1-x}TiO_3$ [3], in which frustration of the exchange interaction occurs due to random dilution. However, it has also been reported that some dilute systems in which little or no frustration is caused by random dilution have shown SG-like behaviour near the percolation threshold (x_p), such as $Fe_xMg_{1-x}TiO_3$ [4] with $x_p \sim 0.24$ [5] and $Fe_xZn_{1-x}F_2$ [6] with $x_p \sim 0.243$ [7]. In the temperature dependence of the zero-field-cooled magnetization M_{ZFC} of these systems, the cusp-like anomaly which is characteristic of SG systems has been observed. However, the microscopic behaviour of the systems $Fe_{0.20}Mg_{0.80}TiO_3$ and $Fe_{0.26}Zn_{0.74}F_2$, probed by Mössbauer time-scale, was found to be remarkably different from that of ordinary SG systems [8, 9]. The magnetically broadened spectrum appears to be superposed on the paramagnetic doublet already at a temperature twice as high as the freezing temperature T_f at which the cusp-like anomaly appears in the temperature dependence of M_{ZFC}. As the temperature decreases, the intensity of the magnetic spectrum increases. On the basis of this temperature dependence of the Mössbauer spectrum, the authors have concluded that the antiferromagnetic (AF) clusters are formed at temperatures much higher than T_f, and that they fluctuate rapidly, just like superparamagnetic particles. With decreasing temperature, the fluctuations of the AF clusters slow down gradually, and the characteristic time of the fluctuation becomes comparable to the time-scale of the ^{57}Fe Mössbauer measurement around $2T_f$. From this dynamical aspect, the authors analysed the spectra by applying the stochastic treatment of relaxation phenomena formulated by Blume [10]. In this way, the glassy behaviour of $Fe_{0.2}Mg_{0.8}TiO_3$ and $Fe_{0.26}Zn_{0.74}F_2$ has been explained to be due to cluster-glass (CG) freezing from the Mössbauer study. In this letter, we report the behaviour of the ac susceptibility of a CG system; the dilute antiferromagnet $Fe_xZn_{1-x}F_2$ near and below the percolation threshold.

---

† Present address: The Institute of Physical and Chemical Research (RIKEN), Wako, Saitama 351-0198, Japan.

In particular, we pay attention to the out-of-phase component of the ac susceptibility χ'' which gives important information for understanding the behaviour of the clusters fluctuating dynamically. The non-dilute compound FeF_2 has the rutile-type crystal structure D_{4h}^{14}–$P4/mnm$ [11], and it establishes an AF long-range order below the Néel temperature $T_N = 78.4$ K [12]. The spin easy axis is parallel to the c-axis. The single crystals of $x = 0.26$ and 0.10 samples used in this study were grown at the University of California, Santa Barbara.

The single crystals of $x = 0.26$ and 0.10 were cut into the form of a parallelepiped of size $2\times2\times4$ mm^3, with its longest axis aligned with the crystalline c-axis. The freezing temperature T_f determined by the cusp temperature in the M_{ZFC}–T curve was 9.7 K and 5.0 K for the $x = 0.26$ and 0.10 samples, respectively. The ac susceptibility was measured using a Quantum Design MPMS5 SQUID magnetometer. The frequency f was varied in the range 1 Hz $\leqslant f \leqslant$1 kHz. The dc and ac magnetic field was applied parallel to the c-axis. The measurements were carried out in an ac magnetic field of 3 G and a bias dc magnetic field, $H_{dc} = 0 \sim 10$ kG, on heating after cooling the sample to $T = 2$ K in zero field.

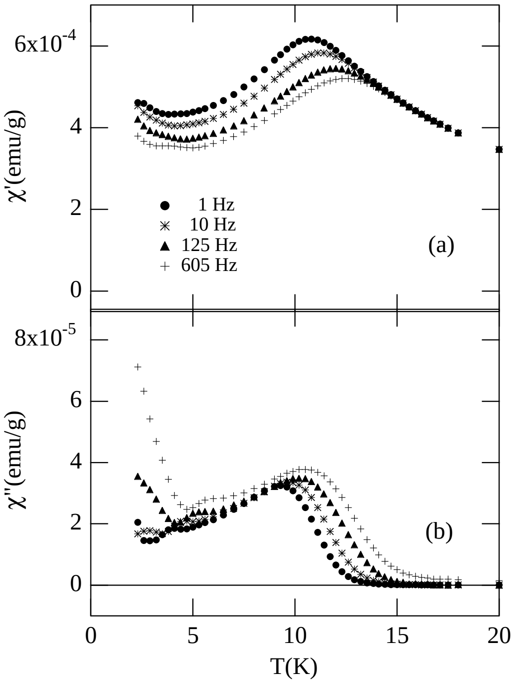

Figure 1. Frequency dependence of the ac susceptibility of $Fe_{0.26}Zn_{0.74}F_2$ obtained under $H_{dc} = 0$. (a) In-phase component χ' and (b) out-of-phase component χ''.

We measured how the temperature dependence of the ac susceptibility of $Fe_{0.26}Zn_{0.74}F_2$ varied with frequency. Typical examples measured in $H_{dc} = 0$ are shown in figure 1. Figure 1(a) shows the in-phase component χ' versus T, while figure 1(b) shows the out-of-phase component χ'' versus T. As is seen in figure 1(a), the χ'–T curve shows a peak whose temperature depends on frequency. This peak of χ' is accompanied by the appearance of the out-of-phase component χ'', as seen in figure 1(b). These behaviours are similar to those of ordinary SG systems qualitatively [1], and very like those of the ac susceptibility of $Fe_{0.25}Zn_{0.75}F_2$ in the temperature range $T > 5$ K reported by Jonason *et al* [13]. When the temperature decreases further, χ' tends to flatten out and turn upward. As pointed out by Jonason *et al* [13], this behaviour indicates the existence of the isolated Fe spins showing a Curie-like behaviour at low temperatures. A similar behaviour has also been observed in some SG systems [14]. However, the temperature dependence of χ'' of $Fe_{0.26}Zn_{0.74}F_2$ at low temperatures is quite peculiar. At a low frequency such as $f = 1$ Hz, χ'' decreases with decreasing temperature. When the frequency increases, χ'' increases suddenly at a certain low temperature. This behaviour indicates that the system is unable to follow the ac field as the frequency increases. Furthermore, there are some irregularities in the temperature dependence of χ'' at $T < 8$ K. As far as we know, such behaviour of χ'' as we observed in $Fe_{0.26}Zn_{0.74}F_2$ has never been reported in ordinary SG systems. It is worth noting that the behaviour of χ'', which reflects the dynamical behaviour, is anomalous in the CG system $Fe_{0.26}Zn_{0.74}F_2$. We therefore pay attention to the irregularities in the temperature dependence of χ'' of $Fe_{0.26}Zn_{0.74}F_2$ in a low temperature region.

We investigated the dc field dependence of the behaviour of the ac susceptibility of $Fe_{0.26}Zn_{0.74}F_2$. The field dependence of the behaviour of χ' of $Fe_{0.26}Zn_{0.74}F_2$ is very similar to that of $Fe_{0.25}Zn_{0.75}F_2$ reported by Jonason *et al* [13]. That is, the peak of the χ'–T curve shifts to low temperatures as the dc field increases similarly to that of ordinary SG systems. However, there is no tendency for the χ''–T curves observed in different dc fields to intersect at low temperatures, which is in contrast to the case of ordinary SG systems [15]. In figure 2, we show a typical example of the bias dc field dependence of the behaviour of the χ''–T curve observed with $f = 125$ Hz. For example, the χ''–T curve measured in zero dc field gives a round peak near 10 K. On the lower temperature side of this main peak, χ'' decreases gently with decreasing temperature and becomes independent of temperature near 6 K. On further cooling, χ'' decreases again with decreasing temperature below the temperature at which the first irregularity appears, $T_1(125\text{ Hz}) = 5.1$ K, and rebounds at the temperature of the second irregularity, $T_2(125\text{ Hz}) = 4.3$ K. The main broad peak which appears associated with the peak of χ' shifts to low temperatures with increasing H_{dc}. Surprisingly, however, the temperatures at which the irregularity appears, T_1 and T_2, do not depend on H_{dc}. We found that T_1 and T_2 do not depend on H_{dc} at all frequencies investigated within our experimental accuracy.

Next we compare the behaviour of the ac susceptibility of $Fe_{0.26}Zn_{0.74}F_2$ ($T_f = 9.7$ K) with that of $Fe_{0.10}Zn_{0.90}F_2$ ($T_f = 5.0$ K), which has also been interpreted as a CG system from the results of the magnetization and Mössbauer measurements [16]. The comparison between the ac susceptibility of $Fe_{0.26}Zn_{0.74}F_2$ and $Fe_{0.10}Zn_{0.90}F_2$ measured with $f = 125$ Hz in $H_{dc} = 0$ is shown in figure 3 as an example. Figure 3(a) shows the in-phase component χ' versus T, while figure 3(b) shows the out-of-phase component χ'' versus T. The peak appears in the temperature dependence of χ' of both samples. The temperature giving the peak of the χ'–T curve of $Fe_{0.26}Zn_{0.74}F_2$ is obviously higher than that of $Fe_{0.10}Zn_{0.90}F_2$. It is natural to consider that the difference in the peak temperature of the χ'–T curve in the two samples originates in the fact that the freezing temperature of $Fe_{0.26}Zn_{0.74}F_2$ is higher than that of $Fe_{0.10}Zn_{0.90}F_2$ by about 5 K. On the other hand, the concentration dependence of χ'' of $Fe_xZn_{1-x}F_2$ is not simple to understand. At temperatures much higher than T_f, the value of χ'' is zero in both samples. When the temperature decreases, χ'' begins to increase in both samples. The onset temperature of the increase in χ'' for $Fe_{0.26}Zn_{0.74}F_2$ is higher than that for $Fe_{0.10}Zn_{0.90}F_2$. This difference of the temperature at which χ'' begins to increase also originates in the difference between the freezing temperatures of both samples. We now pay attention to the irregularities of the χ''–T curve in the low temperature region. As seen in figure 3, the temperatures at which the irregularity appears for $Fe_{0.26}Zn_{0.74}F_2$ are the same

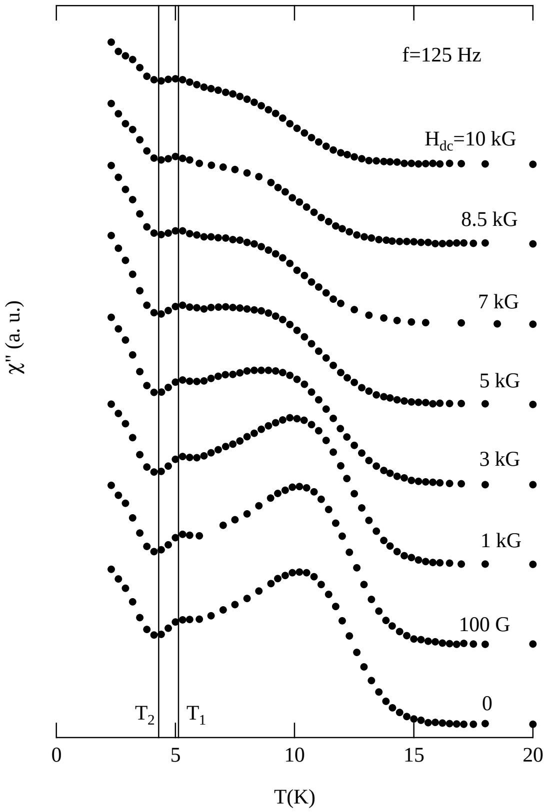

Figure 2. Bias dc field dependence of the out-of-phase component χ'' observed with $f = 125$ Hz in $Fe_{0.26}Zn_{0.74}F_2$. To make the field dependence of the χ''–T curve easy to see, the vertical axis is shifted at each bias dc field. The solid vertical lines indicate the irregularity temperatures T_1 and T_2.

as those for $Fe_{0.10}Zn_{0.90}F_2$. We found that T_1 and T_2 do not depend on the Fe concentration x at each frequency investigated within our experimental accuracy. It is amazing that T_1 and T_2 never change with dilution, although the freezing temperature decreases with decreasing x. This fact suggests that the phenomena accompanying irregularities in the χ''–T curve of $Fe_xZn_{1-x}F_2$ with $x = 0.26$ and 0.10 have no direct relation to the cooperative-phenomenon-like freezing. We also made sure that, for $Fe_{0.10}Zn_{0.90}F_2$, T_1 and T_2 do not depend on H_{dc} at all frequencies investigated.

The log–log plot of T_1 and T_2 as a function of frequency is shown in figure 4. T_1 and T_2 increase linearly with increasing f on the log–log plot. That is, the frequency dependence of T_1 and T_2 is described by a simple power function

$$T_{1,2} = af^b \tag{1}$$

where a and b are constants. We obtained the power b to be 0.05 and 0.07 for T_1 and T_2 respectively.

In the remainder of this letter, we try to discuss the mechanism of the interesting behaviour of χ'' in the CG system $Fe_xZn_{1-x}F_2$ with $x = 0.26$ and 0.10. A good guide for understanding the behaviour of χ'' of $Fe_xZn_{1-x}F_2$, is the study of χ'' of very dilute $Eu_xSr_{1-x}S$ with $x \leqslant 0.10$ that has been reported by Eiselt *et al* [17]. The dilute Heisenberg

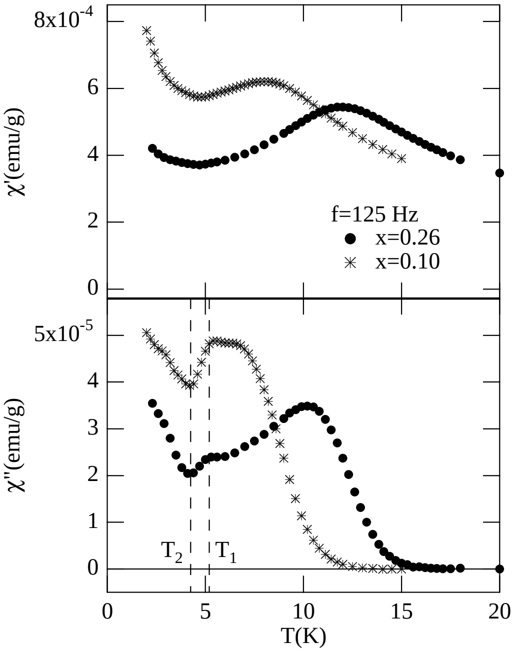

Figure 3. Concentration dependence of the ac susceptibility of $Fe_xZn_{1-x}F_2$ with $x = 0.26$ and 0.10 observed with $f = 125$ Hz under $H_{dc} = 0$. (a) In-phase component χ' and (b) out-of-phase component χ''. The broken vertical lines in (b) indicate the irregularity temperatures T_1 and T_2.

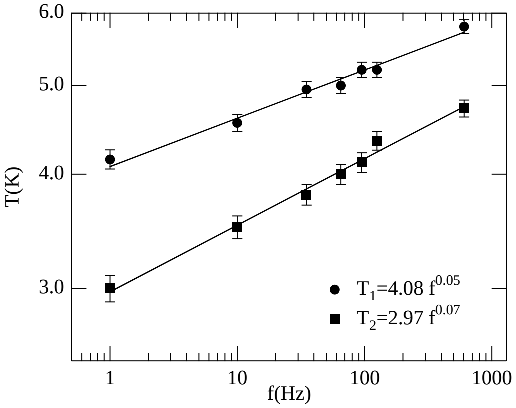

Figure 4. Log–log plot of the temperatures, T_1 and T_2, at which the irregularities appear in the out-of-phase component χ'' of $Fe_xZn_{1-x}F_2$ with $x = 0.26$ and 0.10 as a function of frequency.

ferromagnet $Eu_xSr_{1-x}S$ ($x_p \sim 0.136$ [18]) has been known to behave like the SG system in a wide concentration region $0.1 \leqslant x \leqslant 0.5$ from the low field dc magnetization and ac susceptibility measurements [2]. From careful comparison of the temperature dependence of the Mössbauer spectrum of $Eu_{0.5}Sr_{0.5}S$ and $Eu_{0.1}Sr_{0.9}S$ [19], we believe that the former is the SG system and the latter is the CG one. Eiselt *et al* have reported that a small spike exists on the low temperature side of the main peak in the out-of-phase component of the ac susceptibility of the samples in a very dilute region ($x \leqslant x_p$). They have also interpreted the small spike as being attributable to the contribution from the three-spin cluster by taking into account the result of the theoretical simulations made under the assumption that random small Eu clusters exist. The notable features of the temperature of this small spike are very strong dependence on frequency and independence of Eu concentration. These features are very similar to those of the temperatures at which the irregularities appear in χ'' of the CG system $Fe_xZn_{1-x}F_2$ with $x = 0.26$ and 0.10. The similarity between the microscopic behaviour probed by Mössbauer time-scale and the features of the anomaly in the χ''–T curve between $Eu_xSr_{1-x}S$ and $Fe_xZn_{1-x}F_2$ cannot be ignored, although there are some differences in the circumstances between these two systems, e.g. crystalline field, anisotropy and exchange interaction, etc. Referring to the study of the $Eu_xSr_{1-x}S$ system, we believe that the irregularities of χ'' of $Fe_xZn_{1-x}F_2$ are due to the contribution from the small isolated Fe clusters.

In conclusion, we have made a comprehensive study of the ac susceptibility of the CG system $Fe_xZn_{1-x}F_2$ with $x = 0.26$ and 0.10 in a bias dc field $H_{dc} = 0 \sim 10$ kG. We have found that both samples show irregularities at low temperature in the χ''–T curve. It has been clarified that the irregularity temperatures do not depend on either H_{dc} or Fe concentration x, but do depend strongly on frequency. We interpret this result based on the dynamical random cluster referring to the experimental and theoretical study of the very dilute $Eu_xSr_{1-x}S$ system. We believe that the anomalous behaviour of the χ''–T curve of the dilute system $Fe_xZn_{1-x}F_2$ with $x = 0.26$ and 0.10 gives us important information about the spin dynamics in CG systems.

We would like to thank H Yasuoka and V Jaccarino for supplying the sample which was prepared at UCSB.

References

[1] Fischer K H 1983 *Phys. Status Solidi* b **166** 357
Fischer K H 1985 *Phys. Status Solidi* b **130** 13 and references therein
[2] Maletta H and Felsch W 1979 *Phys. Rev.* B **20** 1245
[3] Tobo A, Ito A and Motoya K 1996 *J. Phys. Soc. Japan* **65** 2249
[4] Kato H, Nakagawa Y, Hosoya S, Kido G, Nakagawa M and Legrand S 1991 *J. Appl. Phys.* **69** 4819
[5] Kato H, Iwai K, Ito A and Nakagawa Y 1995 *J. Magn. Magn. Mater.* **140–144** 1801
[6] Jaccarino J and King J R 1990 *Physica* A **163** 291 and references therein
[7] Sykers M F and Essam J W 1964 *Phys. Rev.* **133** A310
[8] Ito A, Iwai K and Kato H 1995 *J. Phys. Soc. Japan* **64** 1766
[9] Satooka J and Ito A 1997 *J. Phys. Soc. Japan* **66** 784
[10] Blume M 1968 *Phys. Rev.* **174** 351
[11] Stout J W and Reed S A 1954 *J. Am. Chem. Soc.* **76** 6279
[12] Stout J W and Catalano E 1955 *J. Chem. Phys.* **23** 2013
[13] Jonason K, Djurberg C and Nordblad P 1997 *Phys. Rev.* B **56** 5404
[14] Zhou Y, Rigaux C, Mycielski A, Menant M and Bontemps N 1989 *Phys. Rev.* B **40** 8111
[15] Mattsson J, Jonsson T, Nordblad P, Aruga Katori H and Ito A 1995 *Phys. Rev. Lett.* **74** 4305
[16] Satooka J 1998 *PhD Thesis* Ochanomizu University (unpublished)
[17] Eiselt G, Kötzler J, Maletta H and Binder K 1979 *J. Magn. Magn. Mater.* **146–148** 146
[18] Dalton N W, Domb C and Sykers M 1964 *Proc. R. Soc. London.* **83** 496
[19] Maletta H and Crecelius G 1976 *J. Physique* **37** C6 645