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1992 J. Phys.: Condens. Matter 4 L281

(http://iopscience.iop.org/0953-8984/4/16/006)

J. Phys.: Condens. Matter 4 (1992) L281–L286. Printed in the UK

LETTER TO THE EDITOR

Anomalous temperature dependence of magnetization in the applied magnetic field of an amorphous ferrimagnetic insulator

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Received 27 January 1992

Abstract. The numerically obtained temperature dependences of magnetizations in applied magnetic fields for amorphous ferrimagnetic insulators, such as amorphous spinel ferrites, are reported on the basis of a simple model consisting of spin-$\frac{1}{2}$ and spin-1 ions with different transverse fields. The results show a number of characteristic features of the thermal variation, depending on the strength of applied magnetic field. In particular, the anomalous sign reversal of sublattice magnetizations and the hysteresis loop associated with it are found at low temperatures for some systems, when the applied field is not so strong.

In the last few decades there has been an increasing amount of work dealing with magnetic properties of amorphous solids [1, 2]. In particular, many attempts have been made in the past to synthesize ferrimagnetically ordered amorphous ferrites, since they are considered to be technologically important materials. Excepting the work of Sugimoto and Hiramatsu on phosphate glasses [3], these attempts have not succeeded in preparing magnetically ordered amorphous ferrites. Very recently, on the other hand, Srinivasan *et al* [4] have reported that a long-range ferrimagnetically ordered amorphous ferrite is possible in copper ferrite and the saturation magnetization has an order of magnitude larger than the values in [3].

The outstanding feature observed in common in the two works [3, 4] is the temperature dependence of magnetization, which is at variance with that of other crystalline (or amorphous) ferrimagnetic alloys; the magnetization increases markedly in the temperature range below a characteristic temperature T^* ($T^* \simeq 80$ K for amorphous ferrites in phosphate glasses and $T^* \simeq 50$ K for amorphous copper ferrite), although it shows features somewhat similar to those of ferrimagnetic materials in the temperature range above T^*.

In a previous work [5], Kaneyoshi has examined the magnetic structure for amorphous spinel ferrites assuming a two-dimensional even-membered amorphous lattice consisting of spin-$\frac{1}{2}$ and spin-1 ions. The Hamiltonian of the system is then given by

$$H = \sum_{ij} J_{ij} \mu_i^z S_j^z - \Omega_0 \sum_i \mu_i^x - \Omega_1 \sum_i S_j^x \tag{1}$$

where μ_i^α and S_j^α ($\alpha = x, z$) are components of spin-$\frac{1}{2}$ and spin-1 operators at sites i and j. J_{ij} is the nearest-neighbour superexchange interaction assumed to be randomly distributed in strength. Ω_0 and Ω_1 are transverse fields, which are introduced to predict a collinear or canted ferrimagnetic structure, depending on the strength and degree of randomness in J_{ij}. The work shows features similar to the experimental data observed in [3, 4]; one of the present authors (K) has argued that the initial decrease in the magnetization with increasing T arises when weakly coupled spins become paramagnetic due to thermal agitation.

The purpose of this work is to predict the thermal behaviour of magnetization in amorphous spinel ferrites, when a magnetic field is applied. Therefore, the Hamiltonian of the present system is here given by

$$H = \sum_{ij} J_{ij} \mu_i^z S_j^z - \Omega_0 \sum_i \mu_i^x - \Omega_1 \sum_j S_j^x - H\Big(\sum_i \mu_i^z + \sum_j S_j^z\Big). \tag{2}$$

The formulation applied in this work is the same as that in [5], except that the functions $f(x)$, $F(x)$ and $H(x)$ for the magnetizations σ_z, m_z and the quadrupolar moment q_z in [5] are replaced by the new ones: $f(x+H)$, $F(x+H)$ and $H(x+H)$. Here,

$$\sigma_z = \langle\langle \mu_i^z \rangle\rangle_r \qquad m_z = \langle\langle S_i^z \rangle\rangle_r \qquad q_z = \langle\langle (s_i^z)^2 \rangle\rangle_r \tag{3}$$

where $\langle \ldots \rangle_r$ denotes the random bond average. As discussed in [6], the framework is equivalent to the Zernike approximation in the spin-$\frac{1}{2}$ Ising model, which is superior to the standard mean field theory. We report a number of outstanding features of the magnetizations under an applied magnetic field.

Now, in order to evaluate the magnetizations, it is necessary to provide the appropriate form of the probability distribution function $P(J_{ij})$, describing the structural disorder in a simple way. Let us also apply the same function as that in [5];

$$P(J_{ij}) = \tfrac{1}{2}[\delta(J_{ij} - J - \Delta J) + \delta(J_{ij} - J + \Delta J)] \tag{4}$$

with

$$\delta = \Delta J / J \tag{5}$$

where δ is a dimensionless parameter which measures the fluctuation of the exchange interaction. Within the present framework, the distribution (4) is equivalent to the use of the so-called 'lattice model' of amorphous magnets [7]. As discussed in [5], both magnetizations in the sublattices are randomly tilted, like sperimagnets, because of the disorder of J_{ij}, even if the values of Ω_0 and Ω_1 are fixed. The total longitudinal magnetization is then given by

$$\bar{M}_z = (N/2)(\sigma_z + m_z) \qquad \text{and } M_z = \bar{M}_z / N \tag{6}$$

where N is the total number of magnetic atoms.

In figure 1 we show the same results as those (figure 5 in [5]) for $H = 0$, selecting the three values of δ ($\delta = 0.7$, $\delta = 0.9$, $\delta = 0.99$) when the values of Ω_0 and Ω_1 are fixed as $\Omega_0 = 0.01J$ and $\Omega_1 = 1.5J$. These curves are obtained by solving the coupled equations of σ_z, m_z and q_z numerically for the system with a coordination number $z = 4$. The dashed line labelled $\delta = 0.99$ is obtained for the collinear

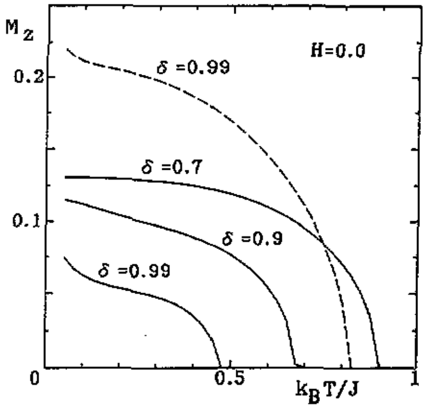

Figure 1. Temperature dependence of M_z (solid curves) in zero magnetic field of the amorphous ferrimagnetic insulator with a coordination number $z = 4$, when the value of δ is changed as $\delta = 0.7$, $\delta = 0.9$ or $\delta = 0.99$. The values of Ω_0 and Ω_1 are then fixed as $\Omega_0 = 0.01J$ and $\Omega_1 = 1.5J$. The dashed curve labelled $\delta = 0.99$ is obtained for the collinear ferrimagnetic system with $\Omega_0 = \Omega_1 = 0$.

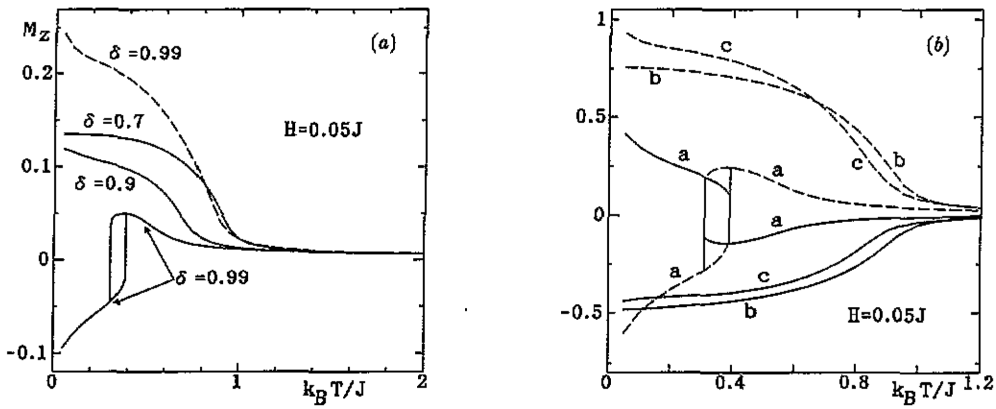

Figure 2. (*a*) Temperature variations of M_z in the weak applied field ($H = 0.05J$) for the same systems as those shown in figure 1. (*b*) Temperature dependences of sublattice magnetizations (σ_z and m_z) in the weak applied field ($H = 0.05J$) for the three systems in (*a*); the systems with $\delta = 0.9$ and $\delta = 0.99$ when the values of Ω_0 and Ω_1 are fixed as $\Omega_0 = 0.01J$ and $\Omega_1 = 1.5J$, and the collinear ($\Omega_0 = \Omega_1 = 0$) system with $\delta = 0.99$. The solid and dashed lines denote σ_z and m_z respectively. Notice that at low temperatures the sign reversal of sublattice magnetizations and the hysteresis loop associated with it are observed for the system (curves *a*) with $\delta = 0.99$, $\Omega_0 = 0.01J$ and $\Omega_1 = 1.5J$. The solid and dashed lines labelled *b* and *c* are for the system (curves *b*) with $\delta = 0.9$, $\Omega_1 = 0.01J$ and $\Omega_1 = 1.5J$ and for the system (curves *c*) with $\delta = 0.99$ and $\Omega_0 = \Omega_1 = 0.0$.

ferrimagnetic system with $\Omega_0 = \Omega_1 = 0.0$. Notice here that the value of M_z at or near $T = 0$ K cannot be determined because of the large numerical errors, although the theoretical estimation for $\Omega_0 = \Omega_1 = 0.0$ may give $M_z = 0.25$ at $T = 0$ K. Thus, the estimated variation of M_z with T for the canted or collinear system with $\delta = 0.99$ shows features similar to the experimental data in [3, 4]; the initial decrease with increasing T arises when weakly coupled spins become paramagnetic due to thermal agitation, since the system includes two exchange interactions $J_{ij} = 1.99J$ and $J_{ij} = 0.01J$ with equal probability.

We now report in the following that the results of figure 1 for $H = 0.0$ may dramatically change, depending on the magnitude of H ($H = 0.05J$ to $H = 2.0J$). Figure 2(a) shows the thermal variation of M_z, when the weak field ($H = 0.05J$) is applied. Comparing the results with those in figure 1, the dashed line with $\delta = 0.99$ and solid lines with $\delta = 0.7$ and $\delta = 0.9$ exhibit essentially similar behaviours, except that for the high-temperature region they take a small but finite value due to the non-zero applied field. For the solid curve with $\delta = 0.99$, however, the features of M_z change drastically; the curve may show the small hysteresis loop in the region near $k_BT/J = 0.4$, which implies that the observable quantity $|M_z|$ may be unstable in the region. This results from the anomalous thermal dependences of σ_z and m_z. For clarification, their variations are depicted in figure 2(b) by selecting the three systems (the solid lines with $\delta = 0.9$ and $\delta = 0.99$, and the dashed line with $\delta = 0.99$ in figure 2(a)). As is seen from figure 2(b), the results of σ_z and m_z for the system with $\delta = 0.99$, $\Omega_0 = 0.01J$ and $\Omega_1 = 1.5J$ (solid and dashed lines labelled a) show the thermal variation to be completely different from that of the other two systems (or the curves labelled b and c). In particular, the anomalous sign reversal of the sublattice magnetizations and the hysteresis loop associated with it are observed for the system (or the curves labelled a) at low temperatures. The existence of a hysteresis loop in the region indicates that the sign reversal of sublattice magnetizations may occur as the first-order transition.

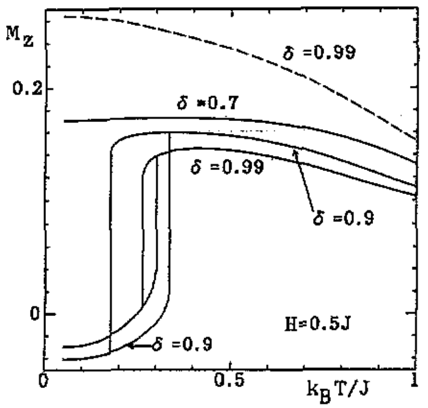

Figure 3. Temperature dependences of M_z in the applied magnetic field ($H = 0.5J$) for the same systems as those shown in figure 1. The same sign reversal of sublattice magnetizations as that (the curve with $\delta = 0.99$, $\Omega_0 = 0.01J$ and $\Omega_1 = 1.5J$) shown in figure 2(b) is also found at low temperatures for the solid lines labelled $\delta = 0.9$ and $\delta = 0.99$.

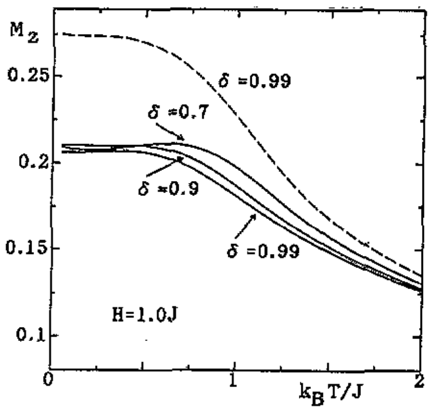

Figure 4. Temperature variations of M_z in the applied magnetic field ($H = 1.0J$) for the same systems as those in figure 1.

Figure 3 shows the thermal dependences of M_z for the same systems as shown in figure 1, when the applied field is given by $H = 0.5J$. As is seen from the figure, the solid lines for $\delta = 0.9$ and $\delta = 0.99$ have a form similar to the solid line with $\delta = 0.99$ in figure 2(a) and hence the $|M_z|$ becomes unstable in the region exhibiting the hysteresis loop. The results found for the solid lines labelled $\delta = 0.9$ and $\delta = 0.99$ also come from the anomalous sign change in the sublattice magnetizations at low temperatures, as is observed for the system (the solid and dashed lines labelled a) with $\delta = 0.99$, $\Omega_0 = 0.01J$ and $\Omega_1 = 1.5J$ in figure 2(b).

For higher applied fields, the thermal variation of M_z for each system in figure 1 may also display some interesting features although the anomalous sign reversal of sublattice magnetizations cannot be observed. In figure 4, the temperature dependences of M_z are plotted for $H = 1.0J$. In contrast to the results of figures 1–3, M_z exhibits a normal (Q-type) behaviour as is usually observed in ferrimagnetic alloys. As shown in figure 5, on the other hand, the magnetization curves for $H = 1.5J$ express the P-type behaviour which has a broad maximum in the intermediate temperature region. Figure 6 shows the thermal variations of M_z for $H = 2.0J$.

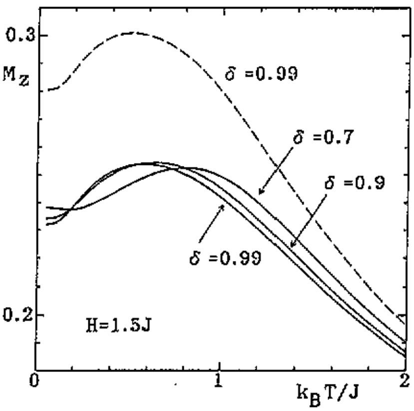

Figure 5. Temperature dependence of M_z in the applied magnetic field $H = 1.5J$ for the same systems as shown in figure 1.

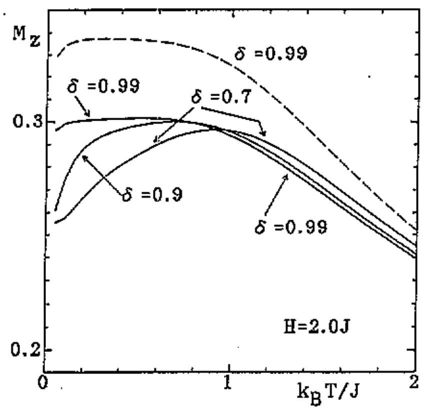

Figure 6. Temperature dependence of M_z in the applied magnetic field $H = 2.0J$ for the same systems as shown in figure 1.

At this point, it is important to note that the sublattice magnetizations σ_z and m_z for figures 4–6 always take negative values for σ_z and positive values for m_z, like the solid and dashed lines in figure 2(b) for the system (curves c) with $\delta = 0.99$ and $\Omega_0 = \Omega_1 = 0.0$ or for the system (curves b) with $\delta = 0.9$, $\Omega_0 = 0.01J$ and $\Omega_1 = 1.5J$. In other words, the characteristic behaviour of M_z in figures 4–6 results from the cancellation between σ_z and m_z, depending on the value of H. In fact, it is necessary to have a rather strong field ($H \simeq 4.0J$) for the sublattice magnetization σ_z to be positive over the whole temperature region. Thus, the solid curves for $\delta = 0.99$ in figure 2(a) and for $\delta = 0.9$ or $\delta = 0.99$ in figure 3 are new results for the thermal variation of magnetization in a ferrimagnetic system; as clearly shown in figure 2(b), they come from the anomalous sign change in the sublattice magnetizations and the hysteresis loop associated with it. The characteristic features essentially depend on the spin configurations at each T resulting from the competition between five values (T, δ, H, Ω_0 and Ω_1), namely the thermal agitation, the strong disorder of J_{ij} ($\delta = 0.9$ or $\delta = 0.99$), the weak applied field ($H = 0.05J$ or $H = 0.5J$), the weak transverse field ($\Omega_0 = 0.01J$) and the strong transverse field ($\Omega_1 = 1.5J$), although, on average, the sublattice magnetizations at each temperature should be non-collinear and take different orientations.

In conclusion, amorphous even-membered ferrimagnetic insulators (such as amorphous copper ferrite [4] and amorphous ferrites in phosphate glasses [3]) may show a variety of new phenomena when the magnetization is measured in an applied magnetic field. In particular, the sign reversal of sublattice magnetizations and the hysteresis loop associated with it may be possible in these systems at low temperatures, when the applied field is not so strong.

References

[1] Kaneyoshi T 1984 *Amorphous Magnetism* (Boca Raton, FL: Chemical Rubber Company)
[2] Moorjani K and Coey J M D 1984 *Magnetic Glasses* (Amsterdam: Elsevier)
[3] Sugimoto M and Hiratsuka N 1982 *Japan J. Appl. Phys.* **21** 197
[4] Srinivasan G, Uma Maheshwar Rao B, Zhao J and Seehra M A 1991 *Appl. Phys. Lett.* **59** 372
[5] Kaneyoshi T 1989 *Solid State Commun.* **69** 91
[6] Kaneyoshi T, Sarmento E F and Fittipaldi I P 1988 *Phys. Rev.* B **38** 2649
[7] Handrich K 1969 *Phys. Status Solidi* **32** K55