Colloque C8, Supplément au n° 12, Tome 49, décembre 1988

EXPANSION OF THE CLUSTER GLASS REGIME BY Fe SUBSTITUTION IN $(Gd_yY_{1-y})(Fe_xAl_{1-x})_2$

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Abstract. – A pronounced increase of the cluster glass regime is observed on the Al rich side for (Gd, Y) $(Fe, Al)_2$, pointing to a competition of ferromagnetic Gd-Gd and antiferromagnetic Gd-Fe exchange interactions. On the iron rich side the magnetic behaviour is dominated by the Fe-Fe interaction. A magnetic phase diagram is proposed.

Although in $Y(Fe_xAl_{1-x})_2$ a concentration of about 78 % Fe is necessary for the appearance of long range magnetic order, the presence of Fe moments even at high dilution is well established. Cluster glass behaviour is observed for $x < 0.78$ [1]. In the Gd-series long range order subsists for all x values and the mean spontaneous Fe moment stays roughly constant [2]. High pressure Mössbauer experiments point to negligible charge transfers between Fe and the substituted Al [3]. A linear decrease of the ordering temperature (T_C) followed for $y \leq 0.22$ by a cluster glass regime is observed in $(Gd_yY_{1-y})Al_2$ [4]. Further informations about the different exchange interactions present in these series were expected from the dilution of the Gd exchange by Y in the pseudo-ternaries $(Gd_yY_{1-y})(Fe_xAl_{1-x})_2$.

Samples with $0 < y < 1$ and $x = 0.05$, 0.1, 0.2, 0.3, 0.7, and 0.8 were prepared, which crystallise in the cubic $MgCu_2$ (C15) structure type. The limited concentration range near $x = 0.5$, where the hexagonal $MgZn_2$ (C14) structure type is stable, was not investigated. Magnetic measurements were performed in fields up to 15 T and in the temperature range 4.2 to 270 K.

On the Al rich side cluster glass behaviour is present for low Gd content. Approaching a critical concentration (y_c) the temperature dependence of the magnetisation measured in low fields becomes complex. Both,

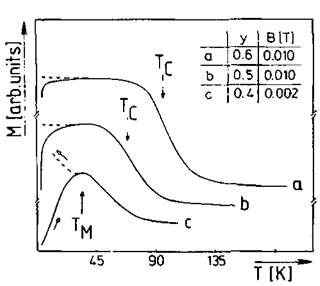

Fig. 1. – Temperature dependence of the magnetisation measured in low fields for $(Gd_yY_{1-y})(Fe_{0.3}Al_{0.7})_2$.

the appearance of long range magnetic order and at low temperatures the existence of a cluster glass is indicated (Figs. 1, 2). A similar behaviour can be observed in $Y(Fe, Al)_2$ near $x = 0.8$ [1]. Above y_c long range order is present. The $M(H)$ data were analysed by $M(H) = M_0(H) + \chi H$. The susceptibility χ exhibits a maximum value of about 8×10^{-5} emu/g near 30-50 at % Gd. The saturation magnetisation (M_S) determined from $M_0(H)$ was found to be lower than the magnetisation of the corresponding Gd sublattice throughout the whole (x, y) range investigated, pointing towards nonvanishing Fe moments and antiferromagnetic Gd-Fe coupling. M_S decreases linearly with decreasing y. Further the slope does not depend on x and is the same as the one obtained for $(Gd, Y)Al_2$. The antiparallel coupling of Gd and Fe is therefore not influenced by the dilution process.

The linear dependence of the freezing temperature (T_M) on the Gd concentration, observed for $(Gd_yY_{1-y})Al_2$ [4], is lost in the Fe containing alloys at least in the explored $y \geq 0.1$ range (inset Fig. 2) and an expansion of the cluster glass region appears. For $x = 0.2$ y_c is about 0.55 compared to $y_c \approx 0.23$ for $(Gd, Y)Al_2$. Further increasing x leads to a decrease of y_c. For $x = 0.7$, $y_c \approx 0.12$ is obtained. In this iron rich concentration range a drastic increase of the ordering temperatures in a small y range is evidenced (Fig. 2). It appears that here long range order involving the strong Fe-Fe interaction is restored by a relatively small amount of Gd.

The deviation from the linear behaviour of $T_C(y)$, observed for $x > 0.1$, indicates an increasing Fe-Fe exchange interaction, although for the Al rich samples the Gd-Gd exchange interaction is the dominant one (Fig. 2). The maximum value of y_c is observed for series with an iron content near that for which a minimum is present in the $T_C(x)$ variation of $Gd(Fe_xAl_{1-x})_2$ [2]. This indicates an alteration in the dominance of the different exchange interactions. In $Gd(Fe, Al)_2$ [5] and in the isostructural series $Gd(Co, Al)_2$ [6] a change in sign of the g-shift is also

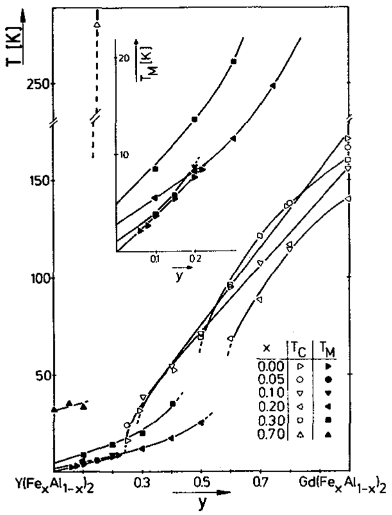

Fig. 2. – Concentration dependence of the ordering temperatures T_C and the freezing temperatures T_M for $(Gd_yY_{1-y})(Fe_xAl_{1-x})_2$. Inset: enlargement of the cluster glass region.

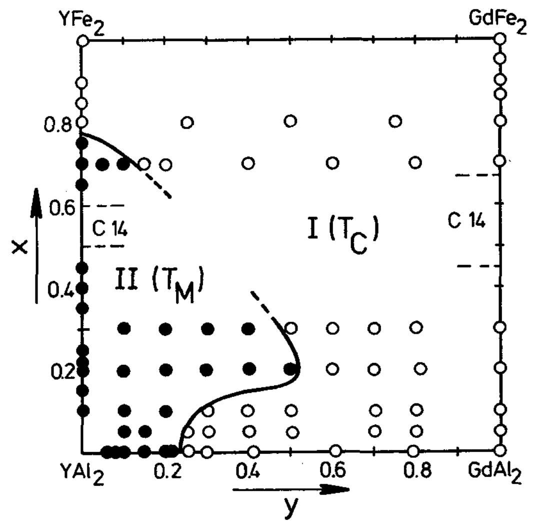

Fig. 3. – Magnetic phase diagram for (Gd, Y) (Fe, Al)$_2$. I: long range ordered (T_Co) region; II: cluster glass (T_M•) region.

reported to occur at about the same 3d-concentration. A possible modification of the character of the conduction band can be inferred from these results, which is also proposed to appear at the Al sites with the onset of magnetic order in $GdAl_2$ [7].

The magnetic phase diagram can thus be divided into two regions (Fig. 3). Samples belonging to region I exhibit long range magnetic order, whereas cluster glass behaviour is observed for samples belonging to region II. The Fe moments, deduced with the two assumptions of (i) constant Gd moment of 7.1 μ_B (as observed in (Gd, Y)Al_2) and (ii) antiferromagnetic coupling of Gd and Fe, stay nearly constant at 1.5 μ_B for all samples in region I. The (x, y)-dependence of the borderline between these two regions may be explained as follows: On the Al rich side the Fe-Fe interaction is negligible and the magnetic interaction is dominated by the competition between the ferromagnetic Gd-Gd and the antiferromagnetic Gd-Fe exchange interaction. Starting from (Gd, Y) Al_2 the substitution of Fe for Al leads to an increasing importance of the Gd-Fe exchange interaction, thus shifting y_c to higher values. For larger x the Fe-Fe interaction starts progressively to be dominant, resulting in the observed diminution of the cluster glass behaviour.

To sum up, the Gd-Fe interaction seems not to be influenced by the dilution process and the expansion of the cluster glass regime points to a competition of the Gd-Fe and the weak Gd-Gd exchange interaction.

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