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2000 J. Phys.: Condens. Matter 12 4253

(http://iopscience.iop.org/0953-8984/12/18/310)

J. Phys.: Condens. Matter **12** (2000) 4253–4262. Printed in the UK PII: S0953-8984(00)09485-6

A superferromagnetic approach for rapidly quenched $Y_{60}Fe_{30}Al_{10}$ alloys

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Received 11 November 1999, in final form 16 March 2000

Abstract. The structural and magnetic properties of $Y_{60}Fe_{30}Al_{10}$ melt-spun ribbons have been studied in this work. The experimental results indicate that $Y_{60}Fe_{30}Al_{10}$ melt-spun ribbons are not homogeneous, i.e. Fe rich clusters are present. The magnetization curves for the ribbons melt spun at 5 and 30 m s^{-1} were analysed with a model based on superferromagnetism. It was found that this superferromagnetic model can be well applied to the ribbon melt spun at 30 m s^{-1}. The Curie transition temperature T_C^{system} was confirmed by the plot of inverse susceptibility versus temperature. For the ribbon melt spun at 5 m s^{-1}, inter-cluster interactions were much stronger and the microstructure was not uniform. Zero-field cooling and field cooling curves showed the cluster behaviour clearly. The Mössbauer results supported the existence of Fe rich clusters and interactions between clusters.

1. Introduction

Recently, magnetic (Nd, Pr)–Fe–(Si, Al) bulk-amorphous materials have shown high coercivity values (Inoue 1995, Inoue *et al* 1997). These materials are not only interesting as a subject for magnetic investigation, but also have potential for many applications, such as permanent magnets, magnetic recording media and magneto-optical devices (Inoue 1995, Inoue *et al* 1997, Li *et al* 1998).

Hard magnetic properties have been reported in binary rare-earth iron alloys prepared by melt spinning (Croat 1982, Croat 1981). It has been found that high coercivities were obtained in ribbons after melt spinning at a relatively low quenching rate (Croat 1982). The coercivity decreased with increasing quenching rate (Croat 1982, Croat 1981). Similar results have been found in melt spun (Nd, Pr)–Fe–(Si, Al) ribbons (Li *et al* 1998, Zhang *et al* 1997). Studies on magnetization and demagnetization processes, magnetic viscosity measurements and Mössbauer investigation suggested the formation of clusters in the amorphous matrix (Inoue 1995, Inoue *et al* 1997, Li *et al* 1998, Yoshiike S *et al* 1998, Ding *et al* 1999, Wang L *et al* 1999, Li *et al* 1999b).

In our previous work, Nd was replaced by the non-magnetic element Y, and $Y_{60}Fe_{30}Al_{10}$ ribbons were prepared at different surface wheel speeds by using a single wheel melt spinner (Li *et al* 1999b). The magnetization curves indicated the presence of Fe rich clusters. However,

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they could not be well fitted with the conventional Langevin function (Li *et al* 1999b, Kneller 1962, Chikazumi 1964). Yoshiike *et al* (1998) have reported that the magnetization curves of rapidly quenched Pr–Fe alloys could be fitted with a modified Langevin function with a Weiss interaction field, probably due to strong interactions between Fe rich clusters. 1–2 nm dark patches were found in the Pr–Fe amorphous phase, supporting the presence of Fe rich clusters. The cluster interactions may result in a ferromagnetic state, which is referred to as super-ferromagnetism (Yoshiike *et al* 1998). In this paper, we have treated the magnetization curves of the melt spun $Y_{60}Fe_{30}Al_{10}$ ribbons using the super-ferromagnetic concept. Since Fe is the only magnetic element in the Y–Fe–Al system, the study can lead to a better understanding of the microstructure of the rare-earth transition metal based amorphous materials.

2. Experiment

The $Y_{60}Fe_{30}Al_{10}$ ingot was prepared by arc-melting of the elements (99.95 pure Y, 99.9% pure Fe and 99.99% pure Al). The ribbons were obtained by melt spinning with a single copper wheel under pure argon atmosphere. The surface speed of the copper wheel was varied between 5 and 30 m s^{-1}.

The details of sample preparation and structural characterization have been reported previously (Li *et al* 1999b). The ribbon melt spun at 30 m s^{-1} was fully amorphous under x-ray diffraction examination. The ribbon melt spun at 5 m s^{-1} consisted of a mixture of an amorphous phase and crystalline Y; no other Y–Fe–Al phases were found. The ^{57}Fe Mössbauer studies at room temperature confirmed that the sample contained a ternary Y–Fe–Al amorphous phase and crystalline Y (Li *et al* 1999b).

In this work, we performed magnetic and Mössbauer studies on the two ribbons melt spun at 5 and 30 m s^{-1}. Magnetic measurements were performed using an Oxford superconducting vibrating sample magnetometer (VSM) with a maximum applied field of 7162 kA m^{-1} (9 T) in the temperature range from 4.2 K to room temperature (RT). Ribbons were investigated by Mössbauer spectroscopy from 4.2 to 290 K. The magnetization was measured in units of A m^2 kg^{-1} (magnetic moment per unit weight). A density of 5.17×10^3 kg m^{-3} was used for the evaluation of magnetic parameters. The density was the average density calculated from the Y, Fe and Al weight percentages in the composition. Our previous works have shown that the average density agreed well with the measured density of the rapidly quenched alloys (Li *et al* 1998, Ding *et al* 1999, Li *et al* 1999b).

3. Results and discussion

3.1. Superferromagnetism

Figure 1 shows the magnetization curve of ribbon melt spun at 5 and 30 m s^{-1} at different temperature. For the ribbon melt spun at 30 m s^{-1}, the magnetization curves taken at 78, 150 and 290 K were similar to the behaviour of superparamagnetic system. A hysteresis loop and a coercivity of 65 kA m^{-1} were observed at 4.2 K, indicating a 'ferromagnetic state'. For the ribbon melt spun at 5 m s^{-1}, magnetization curves taken in the temperature range of 230 and 290 K could be described as indicating superparamagnetism. Magnetic hysteresis was found at lower temperatures. However, these magnetization curves could not be fitted with the conventional Langevin function for superparamagnetism (Kneller 1962, Chikazumi 1964). Strong cluster interaction may be present (Li *et al* 1999b). In addition, the saturation magnetization M_s of the cluster seemed to be a temperature-dependent function. Therefore, we treated the magnetization curves in figure 1 using a superferromagnetic model, which was successfully applied to a rapidly quenched Fe–Pr alloy (Yoshiike *et al* 1998).

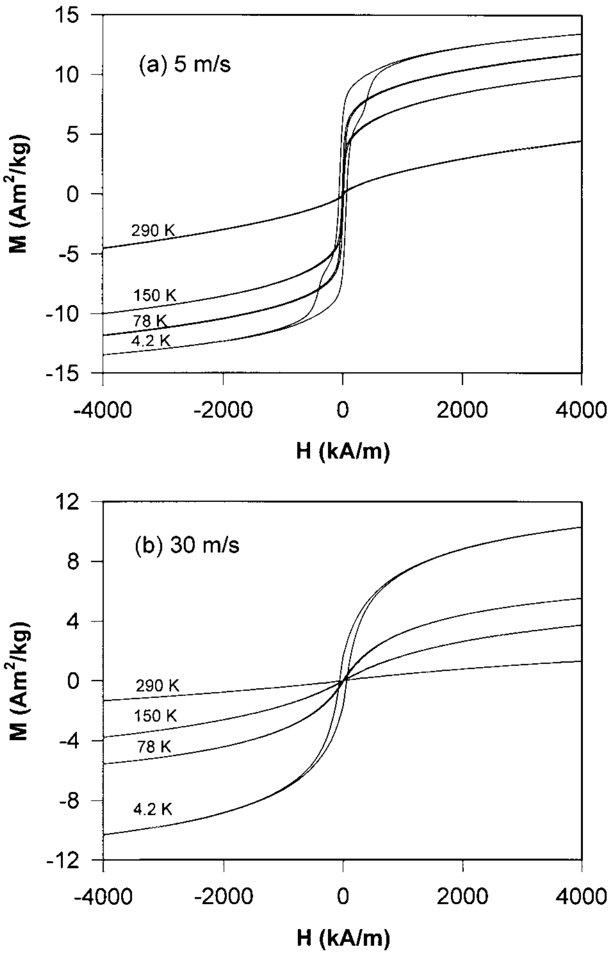

Figure 1. Magnetization curves for the two $Y_{60}Fe_{30}Al_{10}$ ribbons melt spun at (a) 5 m s^{-1} and (b) 30 m s^{-1} at various temperatures.

A system containing small clusters may show a superparamagnetic behaviour. Its magnetization curve above its blocking temperature can be described with a Langevin function as shown below (Kneller 1962, Chikazumi 1964):

$$M = Nm(T)L\left(\frac{m(T)H}{kT}\right). \tag{1}$$

Assuming ferromagnetic coupling between Fe spins within a cluster, $m(T)$ is the spontaneous magnetic moment of a cluster. $m(T)$ is certainly a function of temperature and disappears at the Curie temperature of the cluster phase. $m(T)$ can be expressed as:

$$m(T) = m_s f(T/T_C^{cluster}) \tag{2}$$

where m_s is the saturation moment at zero temperature, and $T_C^{cluster}$ is the Curie temperature of the cluster phase. The temperature dependent function $f(T/T_C^{cluster})$ can be solved from the Brillouin function (Kneller 1962, Chikazumi 1964). In this work, we calculated $f(T/T_C^{cluster})$ assuming the Fe moment $\mu_{Fe} = 2\,\mu_B$, while Y and Al are non-magnetic. From our Mössbauer study, the Fe moment is expected to be in the range of 1–1.5 μ_B, as discussed later. The

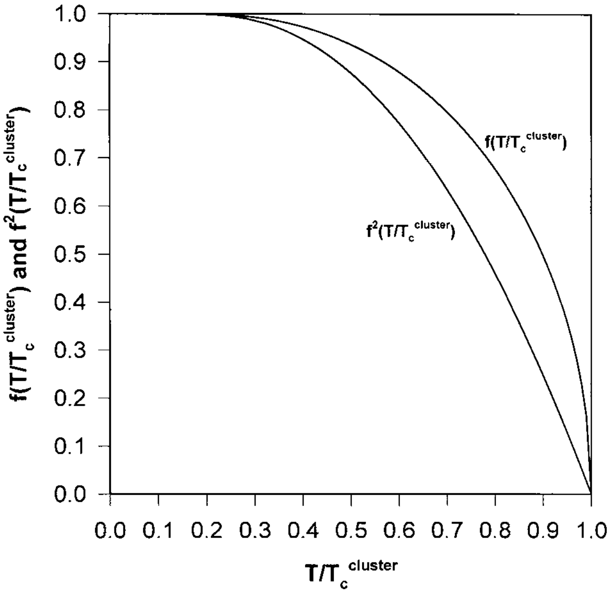

Figure 2. $f(T/T_C^{cluster})$ and $f^2(T/T_C^{cluster})$ versus $T/T_C^{cluster}$ curves.

magnetization $M(T)/M_s$ curves for Fe and Ni are close to the calculated $f(T/T_C)$ based on $\mu_{Fe} = 2\ \mu_B$ (Kneller 1962, Chikazumi 1964).

Between clusters, interactions may be present. To describe the interactions between clusters, the magnetic field H is replaced by the addition of an external magnetic field H and an internal magnetic field (molecular field) αM. The Langevin function is therefore:

$$M(T,H) = Nm_s f\left(\frac{T}{T_C^{cluster}}\right) L\left(\frac{m_s f(T/T_C^{cluster})(H+\alpha M(T,H))}{k_B T}\right). \tag{3}$$

Figure 3 shows the Langevin function fitting curves $(M(T,H)/f(T/T_C^{cluster}))$ versus $f(T/T_C^{cluster})(H+\alpha M)/k_B T)$ for the two ribbons ((a) 5 m s^{-1} and (b) 30 m s^{-1}). For the ribbon melt spun at 30 m s^{-1}, a good fitting result could be obtained in the temperature range from 290 K to 30 K and in the field range from 0 to 7162 kA m^{-1} (0–9 T). $T_C^{cluster}$ was determined to be 360 K, and α was calculated to be 3. This showed that these clusters should contain a significant amount of non-magnetic Y and Al, because the Curie temperature of 360 K is much lower than that of pure Fe ($T_C = 1043$ K). Figure 3(a) shows the fitting result of the ribbon melt spun at 5 m s^{-1}. An acceptable fit is obtained in the temperature range from 290 K to 150 K, in the field range from 0 to 7162 kA m^{-1} (0–9 T). $T_C^{cluster}$ was 580 K and α was 61. $T_C^{cluster}$ of the ribbon melt spun at 5 m s^{-1} is much higher than that of the ribbon melt spun at 30 m s^{-1}. This demonstrates that the interaction between the Fe atoms in clusters in ribbon melt spun at 5 m s^{-1} is stronger than that in ribbon melt spun at 30 m s^{-1}. Interactions between clusters in the ribbon melt spun at 5 m s^{-1} ($\alpha = 61$) were much stronger than that in the ribbon melt spun at 30 m s^{-1} ($\alpha = 3$). In comparison with the α value of 1230 for bcc-iron in the Weiss theory (Kneller 1962, Chikazumi 1964), the interactions between the clusters are still much weaker than the interactions between the Fe spins in the bcc-Fe phase.

Through the cluster interactions (αM), the system (sample) can be transformed from paramagnetic (superparamagnetic) to ferromagnetic. The Curie temperature of the whole

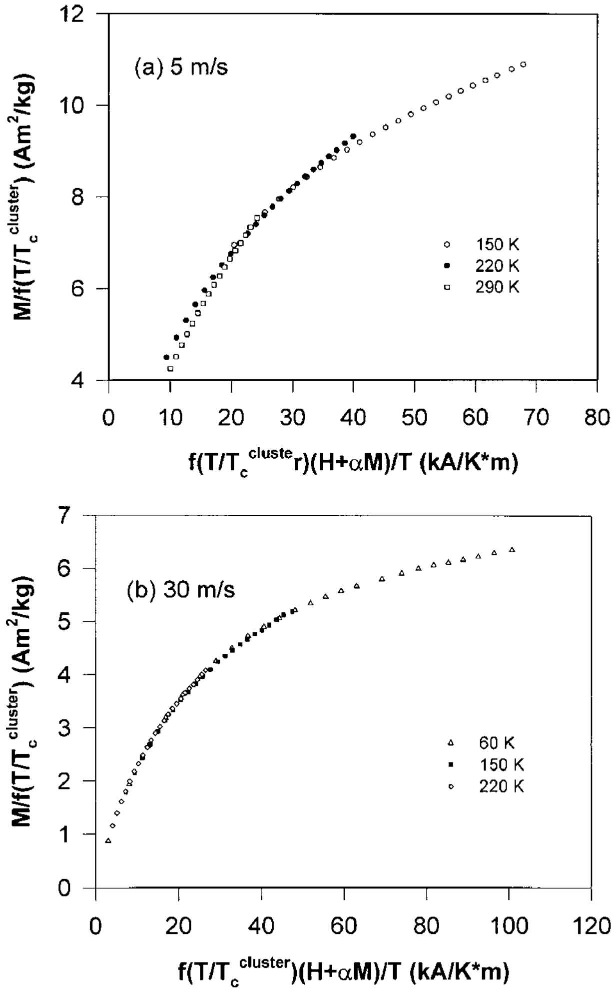

Figure 3. $M/f(T/T_C^{cluster})$ against $f(T/T_C^{cluster})(H+\alpha M)/T$ curves for the $Y_{60}Fe_{30}Al_{10}$ ribbons melt spun at (a) 5 m s^{-1} and (b) 30 m s^{-1}.

system T_C^{system} gives the strength of the magnetic interactions between clusters. T_C^{system} can be estimated from the initial slopes of the universal curves and the α value determined above.

For $x \ll 1$, Langevin function $L(x)$ approximates to $x/3$. Then equation (1) can be expressed as

$$M(T,H) = \frac{Nm_s^2 f^2(T/T_C^{cluster})(H+\alpha M(T,H))}{3k_B T}. \tag{4}$$

The magnetic moment of a cluster is derived as:

$$m_s = \frac{3k_B}{M_0}\frac{M(T,H)T}{f^2(T/T_C^{cluster})(H+\alpha M)} \tag{5}$$

where $M(T,H)T/f^2(T/T_C^{cluster})(H+\alpha M)$ is the initial slope of the fitted Langevin type curve in figure 3. $M_0 = Nm_s$ is the saturation magnetization at 0 K. We used the magnetization value measured at 4.2 K at 7162 kA m^{-1} (9 T) as M_0. At $T = T_C^{system}$ and $H = 0$, using

Table 1. Parameters derived from the analysis of the magnetization data for ribbon melt spun at 5 and 30 m s^{-1}.

	5 m s^{-1}	30 m s^{-1}
Curie temperature in a cluster $T_C^{cluster}$ (K)	500	360
Molecular field coefficient among clusters α	61	3
Average magnetic moment of a cluster $\langle m^2\rangle/\langle m\rangle$ (μ_B)	~600	~500
Number of clusters N (10^{19} cm^{-3})	~6.7	~8
Number of iron atoms in a cluster	~300	~250
Curie temperature for cluster coupling T_C^{system} (K)	135	8

equation (3), T_C^{system} can be obtained as:

$$T_C^{system} = Nms^2 f^2(T_C^{system}/T_C^{cluster})\alpha/3k_B. \tag{6}$$

All the derived parameters are listed in table 1. The Curie temperature of the system (T_C^{system}) for the ribbon melt spun at 5 m s^{-1} is much higher than that for the ribbon melt spun at 30 m s^{-1}, due to much stronger interactions between clusters. The enhancement of cluster interactions may be explained by formation of a large quantity of crystalline yttrium (Li *et al* 1999b), so that the clusters were much more closely packed. Using $\mu_{Fe} = 2\ \mu_B$, the average number of Fe atoms in a cluster can be derived. Compared between the two ribbons melt spun at 5 and 30 m s^{-1}, the clusters in the two samples contain nearly the same number of Fe atoms. However, $T_C^{cluster}$ for the ribbon melt spun at 5 m s^{-1} is 500 K, which is significantly higher than that of 360 K for the ribbon melt spun at 30 m s^{-1}, probably due to a difference in the cluster composition.

3.2. Low-field magnetic behaviour

To determine the region of irreversibility, samples were cooled to 4.2 K in zero applied field (zero-field cooling process—ZFC). Then a constant field (150 Oe) was applied during the temperature increase, and the ZFC magnetization $M_{ZFC}(H)$ was measured up to 250 K. The temperature was then reduced to 4.2 K again and the $M_{FC}(H)$ measured in the same applied field during the cooling (field cooling process—denoted as FC).

Figure 4 shows the ZFC and FC curves for the $Y_{60}Fe_{30}Al_{10}$ sample melt spun at 5 and 30 m s^{-1}. The onset of irreversibility starts before the cusp of the peak suggesting that clusters of spins other than individual spins are involved in the freezing phenomenon, as has been previously observed in other systems with a spin-glass-like behaviour (Li *et al* 1999a).

The sample melt spun at 30 m s^{-1} has a sharp peak in its value of M at around 30 K, while the ribbon melt spun at 5 m s^{-1} has a broad peak in the temperature range 50–100 K. These results indicate that the latter sample has a non-uniform structure (with respect to cluster size or cluster interactions).

From equation (4), the initial susceptibility χ at low field can be expressed as:

$$\chi = Nms^2 f^2(T/T_C^{cluster})/3k_B(T - T_C^{system}). \tag{7}$$

The insets in figure 4 show χ^{-1} versus temperature for the ribbons melt spun at 5 and 30 m s^{-1}. From figure 2, we can see that $f(T/T_C^{cluster})$ and $f^2(T/T_C^{cluster})$ are not strongly dependent on temperature when $T/T_C^{cluster}$ is in the range 0 to 0.4. Therefore, a plot of χ^{-1} versus temperature is expected to be almost a straight line at lower temperatures. For the ribbon melt spun at 30 m s^{-1}, its plot is nearly linear, while for the ribbon melt spun at 5 m s^{-1}, χ^{-1} has a complex temperature dependence.

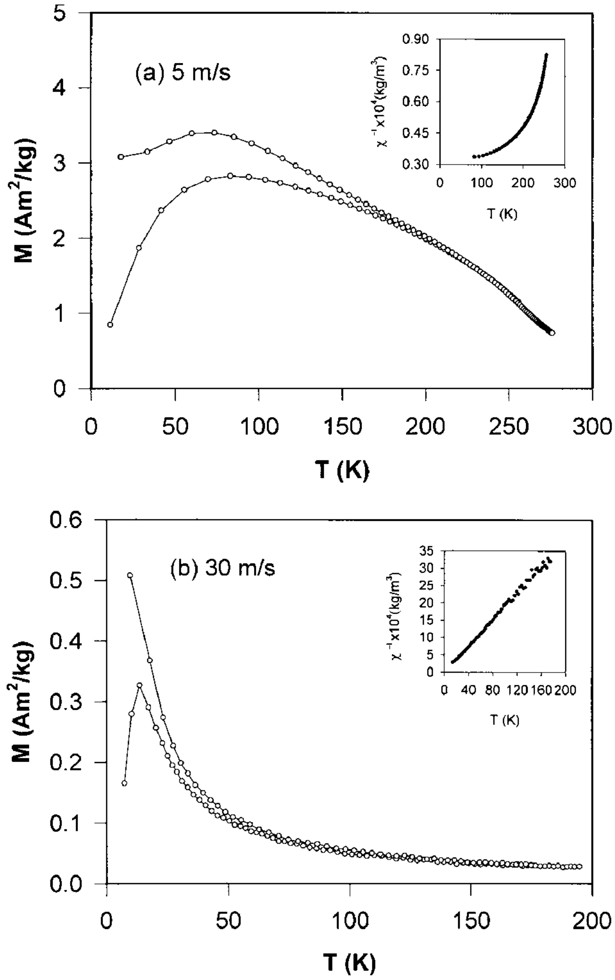

Figure 4. Zero-field cooling (ZFC) and field cooling (FC) curves for the $Y_{60}Fe_{30}Al_{10}$ ribbons melt spun at (a) 5 and (b) 30 m s^{-1}. The applied magnetic field was 150 Oe. Inset: the inverse susceptibility $1/\chi$ taken from the ZFC curve as a function of temperature.

$T_C^{system} = 7 \pm 2$ K for the ribbon melt spun at 30 m s^{-1} can be obtained from the inverse susceptibility plot extrapolated to zero. The T_C^{system} value is nearly identical to that obtained from the superferromagnetic fitting (figure 3 and table 1). This suggests that the superferromagnetic model is suitable for the ribbon melt spun at 30 m s^{-1}. For the ribbon melt spun at 5 m s^{-1}, χ^{-1} versus temperature is approximately linear only at higher temperatures ($\geqslant$240 K). The T_C^{system} obtained for the ribbon melt spun at 5 m s^{-1} is very approximately 220 K by extrapolation of the linear part. This value is higher than that obtained from the superferromagnetic treatment (table 1).

3.3. ^{57}Fe *Mössbauer spectroscopy*

Mössbauer measurement of the ribbons melt spun at 5 and 30 m s^{-1} are shown in figure 5. For the ribbon melt spun at 30 m s^{-1}, only the spectrum taken at 4.2 K has a clear magnetic

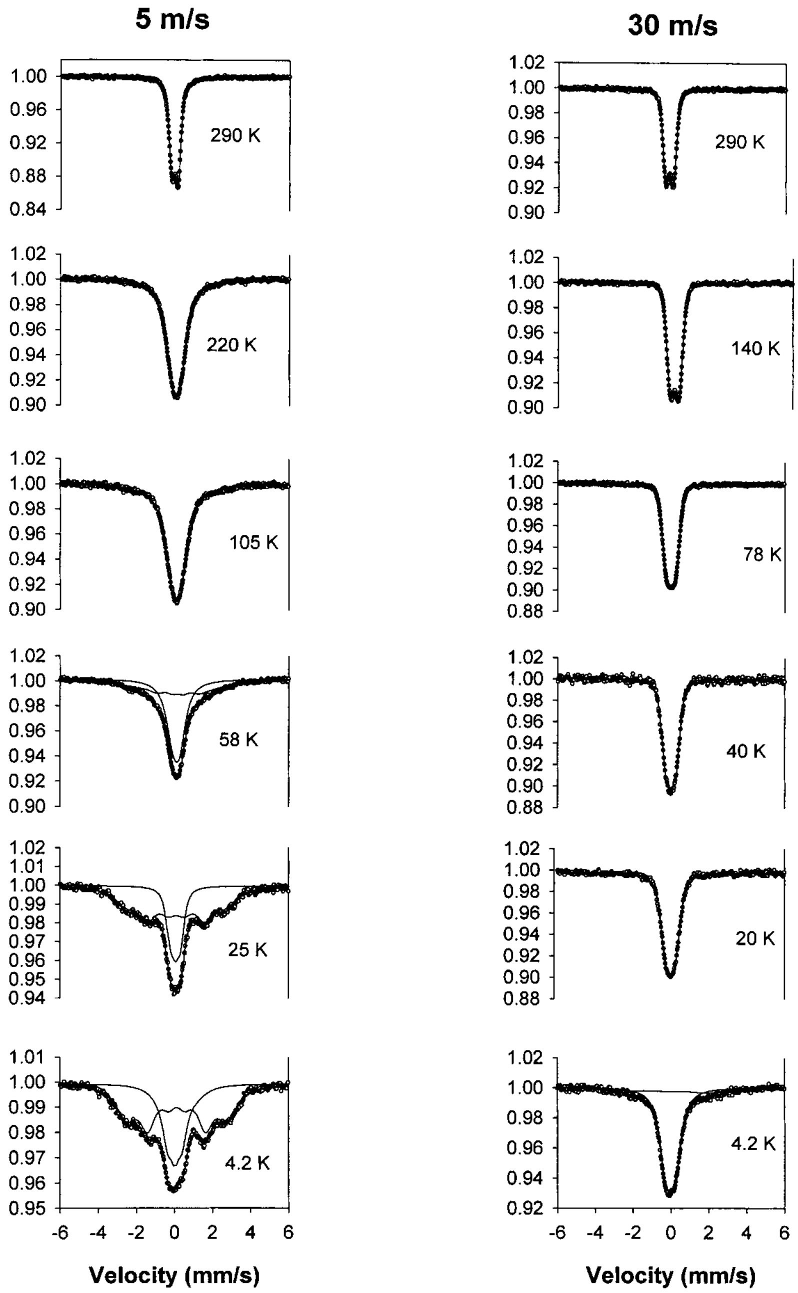

Figure 5. Mössbauer spectra for the $Y_{60}Fe_{30}Al_{10}$ ribbons melt spun at 5 and 30 m s^{-1} at various temperatures.

splitting. This result is in good agreement with the T_C^{system} of 8 K determined from the superferromagnetic treatment. For the ribbon melt spun at 5 m s^{-1}, even the spectrum taken at 220 K has a non-linear baseline, indicating the presence of ferromagnetism. The spectrum taken at 4.2 K consists of a mixture of a paramagnetic part and a ferromagnetic part, indicating inhomogeneity. The average hyperfine field of the ferromagnetic part was 26 T, corresponding to an Fe moment of 1.73 μ_B using the hyperfine field/magnetic moment ratio of 15 T μ_B^{-1}.

We used dynamic lineshape site analysis (Blume and Tjon 1968) to fit our Mössbauer spectra. At higher temperatures, the spectra of ribbon melt spun at 5 m s^{-1} could be well fitted with one site, as shown in figure 5. We could not obtain satisfactory fitting at low temperatures in one site. We fitted the spectra at 58, 25 and 4.2 K in two sites. The site with a higher hyperfine field shows very clear magnetic splitting. The population of the site with a clear magnetic splitting increases with the decrease of temperature. The population of the site with the higher hyperfine field is about 65% at 4.2 K. The average hyperfine field also increases with decreasing temperature. The spectra for ribbon melt spun at 30 m s^{-1} can be fitted in one site except for that at 4.2 K. The site population of the site with a higher hyperfine field is about 20% at 4.2 K.

From these results of Mössbauer measurements, we can find that the superferromagnetic model can be used in the temperature range within which there is no clear magnetic splitting. This indicates that superferromagnetic model is not suitable for systems with relatively strong interactions among clusters.

The Mössbauer analysis demonstrates that the amorphous structure in the Y–Fe–Al ribbons is not homogeneous. The existence of a non-zero average hyperfine field above its blocking temperature (or T_C^{system} in this work) suggests the existence of short-range magnetic order or clusters (Chappert *et al* 1981). The increase in the population of the site with the higher hyperfine field with decreasing temperature can be well described by (i) a relatively broad distribution of the cluster size, (ii) nonuniformity of cluster composition or (iii) cluster interactions.

4. Summary

$Y_{60}Fe_{30}Al_{10}$ alloys were prepared by melt spinning at two different wheel speeds (5 and 30 m s^{-1}). X-ray diffraction and DSC examination (Li *et al* 1999b) showed that the ribbon melt spun at 30 m s^{-1} consisted of an amorphous phase, while a mixture of crystalline yttrium and amorphous phase was found in the ribbon melt spun at 5 m s^{-1}.

The magnetization curves of the ribbon melt spun at 30 m s^{-1} could be well fitted by the superferromagnetic Langevin function. The clusters were calculated to contain an average number of approximately 250 Fe atoms. The whole system could transform to a ferromagnetic state through interactions between the clusters. Zero-field cooling and field cooling curves confirmed the transition. The plot of inverse susceptibility versus temperature could be well fitted by a straight line. The Curie temperature of the system determined from the linear relationship was nearly equal to that determined from the superferromagnetic fitting.

The magnetization curves of ribbon melt spun at 5 m s^{-1} could be approximately fitted only at relatively high temperatures ($\geqslant$150 K). From the fitted parameters, the average number of Fe atoms in a cluster is very close to that for the ribbon melt spun at 30 m s^{-1}. The sample possessed a much higher Curie temperature of the system due to much stronger interactions between clusters. This is probably due to the formation of crystalline yttrium, so that the Fe rich clusters are much more closely packed. The zero-field cooling and field cooling curves and the $1/\chi$ versus T plot indicate that the sample melt spun at 5 m s^{-1} is not uniform and may have a broad distribution of cluster interactions, cluster sizes or cluster compositions (or a combination of these).

Our Mössbauer analysis of the ribbons melt spun at 5 and 30 m s^{-1} at different temperature confirmed the inhomogeneity of the samples, and therefore supported the formation of Fe rich clusters.

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