Dynamic susceptibility of a strong random anisotropy magnet

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Frequency dependent complex ac susceptibility measurements have been made on amorphous $Dy_xY_{7-x}Ni_3$, a model strong random magnetic anisotropy (RMA) system. Analysis of these measurements in terms of coexisting free and correlated or clustered Dy spins has enabled the evolution of the distribution of spin relaxation times to be mapped as a function of temperature from the paramagnetic phase into the low-temperature speromagnetic phase. Qualitatively the results are similar to those observed close to T_g for spin-glass systems, but with a major difference: for the strong RMA *a*-$Dy_xY_{7-x}Ni_3$ system the spin relaxation times, as T_g is approached from above, are of the order of seconds. This is some four orders of magnitude longer than those observed for typical spin glasses. [S0163-1829(96)08145-3]

INTRODUCTION

Magnetic systems possessing random magnetic anisotropy (RMA) have attracted considerable attention over the last decade. The presence of RMA gives rise to interesting magnetic phases which cannot be recognized in systems with high symmetry. An ideal experimental realization of an RMA system is usually achieved in amorphous alloys containing non-*S*-state rare earth ions in a nonmagnetic host. The topological disorder inherent in such amorphous systems gives rise to internal crystalline electric fields which are almost random from rare-earth site to rare-earth site. Single ion anisotropy, therefore, dominates. Many theoretical[1–7] and experimental[8–10] studies have shown that long range magnetic order is not possible in such RMA materials. Instead, the nature of the magnetic ground state is determined by the ratio of the average local anisotropy, D, to the exchange strength, J. For $D/J \ll 1$ Chudnovsky and co-workers[4,5,11] have proposed a magnetic phase described as a correlated speromagnet or a ferromagnet with wandering axis, while in the strong RMA limit, $D/J \geqslant 1$, a speromagnetic state is predicted. The speromagnetic state is often considered as closely analogous to the spin glass state.[3,12] However, analysis of the critical phenomena has led some authors[13] to suggest that the speromagnet belongs to an entirely new universality class.

A meaningful comparison between the spin glass and speromagnetic states cannot be made without recourse to detailed consideration of the temperature-dependent spin dynamics of the two types of system. Surprisingly there have been relatively few studies of spin dynamics in the strong RMA limit. We have therefore carried out a detailed study of the dynamic magnetic susceptibility of a model strong RMA system; the amorphous alloy series, $Dy_xY_{7-x}Ni_3$ with $0<x\leqslant 7$. All the alloys have been previously characterized by neutron diffraction, which indicates[14] a random substitution of Dy by Y over the whole composition range. In addition it is found that that the Ni atoms do not support a moment.[15,16] Our preliminary ac susceptibility studies[15] showed that at all Dy concentrations the alloys are speromagnetic below a glasslike transition temperature T_g, which decreases with decreasing x. The speromagnetic phase has also been verified by our small angle neutron diffraction measurements[17] which show that amorphous $Dy_xY_{7-x}Ni_3$ alloys do not support long range order at any composition. Instead, below T_g the range of the magnetic correlations is limited to the order of one nanometer.

The vital role of single ion random anisotropy in defining the magnetic properties of the amorphous $Dy_xY_{7-x}Ni_3$ compounds is apparent when the magnetic behavior of the structurally similar $Gd_xY_{7-x}Ni_3$ system is considered. For the latter compounds susceptibility and magnetization measurements[15] indicate behavior associated with conventional random exchange systems: the dilute *S-state* Gd ions give rise to a spin glasslike state below $x=3.3$. Above this limit the onset of ferromagnetic order, witnessed by a rapid saturation of the dc magnetization is well described by percolation within a simple Heisenberg statistical nearest neighbor model (with a coordination number of 12). In $Dy_xY_{7-x}Ni_3$ percolation of the non-*S*-state Dy ions is entirely absent and, as we shall show, the spin dynamics are significantly different to those of a conventional, random exchange, spin-glass system.

EXPERIMENTAL TECHNIQUES

A crystalline precursor of each $Dy_xY_{7-x}Ni_3$ sample was prepared in an argon arc furnace using the appropriate atomic percentages of metals of at least $3N$ purity. No sample showed more than a 0.02% mass loss after arc melting. The crystalline ingots were subsequently induction melted and quenched onto a single copper wheel (surface velocity of 70 m/s) to produce amorphous ribbons of approximately 20-μm thick and 1.5-mm wide. X-ray-diffraction measurements of the side of the ribbon closest to the wheel showed the samples to be amorphous. Although x-ray diffraction did reveal a small percentage of crystallinity on the opposite surface of the ribbon the volume fraction of the crystalline impurity phase must be extremely small, as bulk neutron diffraction measurements showed no evidence of crystallinity in any sample.[14]

ac susceptibility measurements were obtained using a modified Hartshorn bridge network. A dual phase lockin amplifier (EG&G 5208) enabled both the in-phase and out-of-

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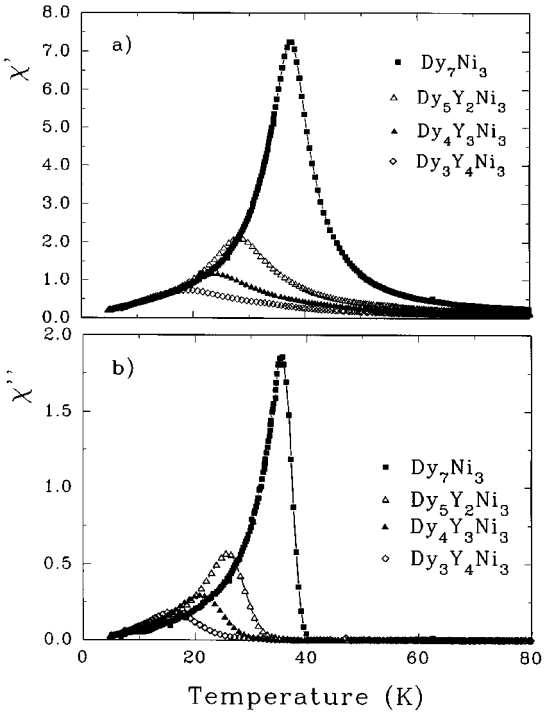

FIG. 1. The real (a) and imaginary (b) components, χ' and χ'', of the ac susceptibility of amorphous $Dy_xY_{7-x}Ni_3$ alloys measured at 330 Hz.

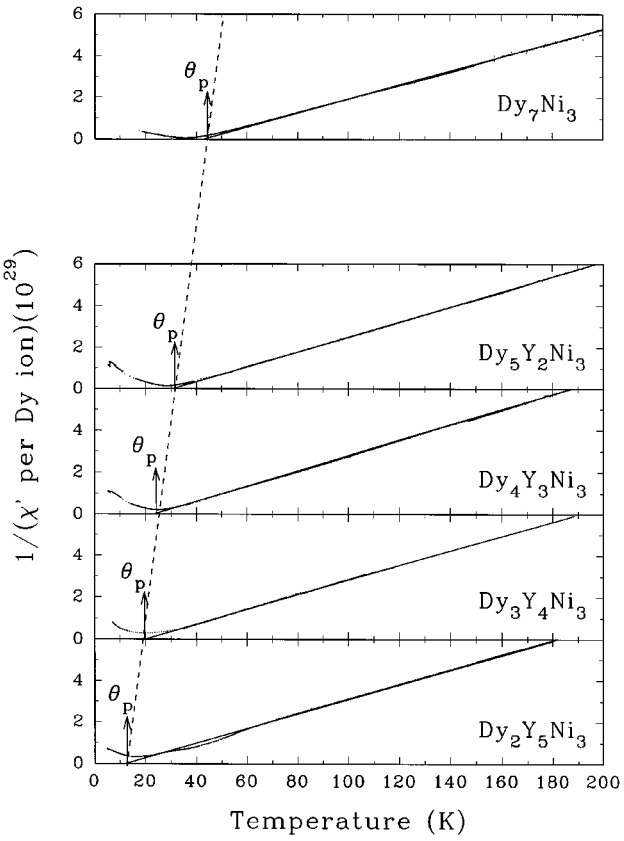

FIG. 2. The inverse real component of susceptibility as a function of temperature for amorphous $Dy_xY_{7-x}Ni_3$ alloys with $x=2$, 3, 4, 5, and 7. The arrows indicate the respective paramagnetic Curie temperatures, θ_p, obtained from a linear extrapolation of the high-temperature data.

phase components to be measured simultaneously. Temperatures in the range $4.2 \leqslant T \leqslant 290$ K were sampled by warming at a steady rate of approximately 0.3 K/min. This relatively slow rate enabled the measuring frequency to be scanned over the range 11 to 5670 Hz at each temperature. The susceptibility measurements were all made in an rms field of 10 μT applied parallel to the length of the amorphous ribbons. The coil design allowed the use of samples in the form of ribbons of up to 5-cm long, thereby reducing demagnetizing factors to less than 2%. The ac susceptometer was calibrated using standard superconducting and ferromagnetic samples with known demagnetizing factors. The susceptibility data presented in this paper have been calibrated according to SI notation and are therefore dimensionless.

ac SUSCEPTIBILITY MEASUREMENTS

Initial characterization of the $Dy_xY_{7-x}Ni_3$ magnetic samples, carried out at 330 Hz, is shown in Fig. 1. A large increase in the magnitude of the real component of the susceptibility, χ', together with a sharpening of the susceptibility cusp and an increase in the cusp temperature T_g is observed with increasing Dy concentration. Below T_g χ' decreases rapidly and is extremely small for $T \ll T_g$. This behavior is typical of speromagnets with strong RMA:[18] at low temperatures the strong single ion anisotropy pins the Dy moments along the local random anisotropy axis and thus restricts the free orientation of each Dy spin in the applied oscillating field.

For temperatures greater than approximately $2T_g$ the susceptibility of all the samples is precisely of the Curie-Weiss form. The gradient of the inverse susceptibility (Fig. 2) is consistent with a Dy moment appropriate to the free ion value of $10.6\mu_B$. The paramagnetic Curie temperature, θ_p, is positive and varies linearly with x. This indicates a predominantly ferromagnetic Dy-Dy exchange with an average exchange strength which increases linearly with Dy concentration. Despite a positive θ_p the presence of antiferromagnetic interactions cannot be entirely precluded. However, we note that our neutron-diffraction measurements[14] indicate strongly damped but predominantly ferromagnetic spin correlations, and also draw attention to the conventional ferromagnetism observed in the analogous amorphous $Gd_xY_{7-x}Ni_3$ system.[15] Deviations from Curie-Weiss behavior are evident as the temperature approaches T_g. As discussed below, we attribute these deviations to the presence of clustering of the Dy spins.

Extension of the above ac susceptibility measurements to a broad frequency range (11–5670 Hz) reveals a marked frequency dependence of χ_{ac}. The magnitude of this dependence is significantly greater than that observed in conventional spin-glass systems and provides an indication that the time scale of the magnetic relaxation close to T_g is commensurate with the time window of the ac measurements. Figures 3(a) and 3(b) show the real component of the susceptibility $\chi'(T,\omega)$ for six different frequencies between 11–5670 Hz for the Dy_7Ni_3 and $Dy_3Y_4Ni_3$ samples respectively. For clarity the out-of-phase component of susceptibility, $\chi''(T,\omega)$ is presented only for measuring frequencies of 11 and 5670 Hz.

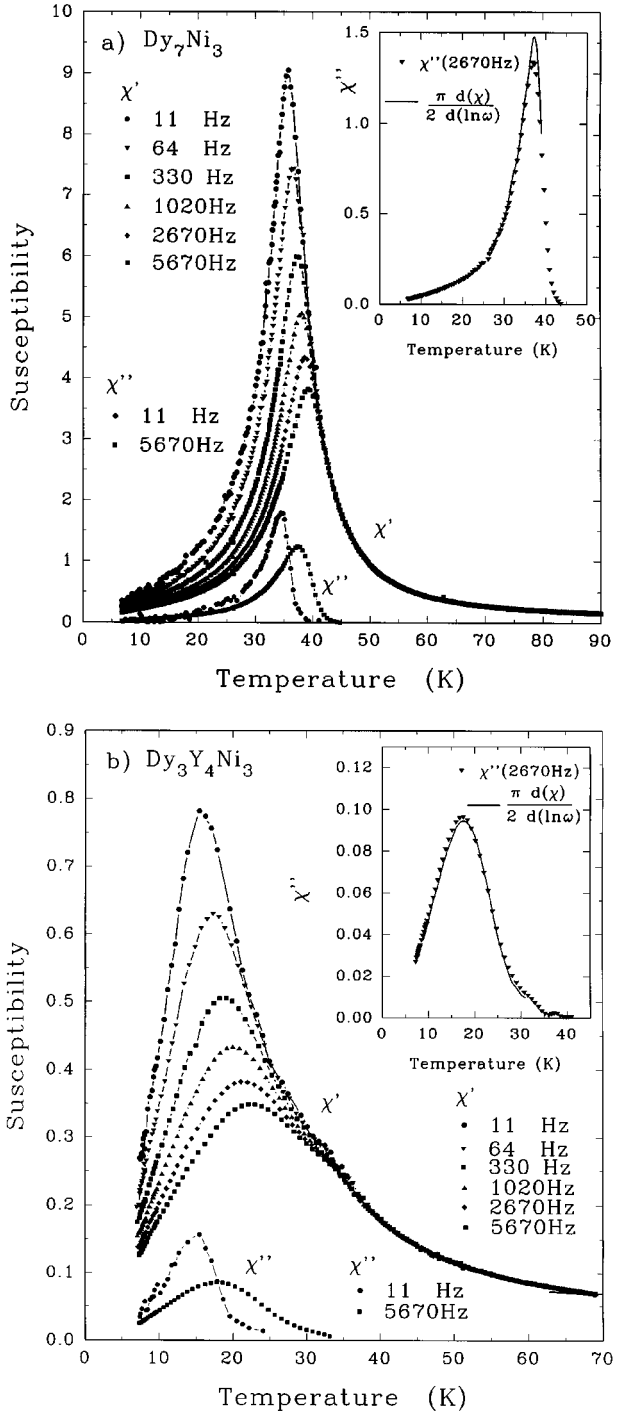

FIG. 3. The frequency dependence of the susceptibility, $\chi'(\omega)$ and $\chi''(\omega)$ of (a) Dy_7Ni_3 and (b) $Dy_3Y_4Ni_3$. The inset in both (a) and (b) shows χ'' measured at 2670 Hz together with the Lundgren relation (solid line) estimated using Eq. (2).

For temperatures much higher than T_g the samples exhibit no frequency dependence in $\chi'(T,\omega)$ within experimental accuracy. This feature, together with the absence of any imaginary component, $\chi''(T,\omega)$, indicates that the measured $\chi'(T,\omega)$ approximates closely to the isothermal susceptibility $\chi(T,0)$. On cooling, however, a significant $\chi''(T,\omega)$ develops, and is first observed at the highest frequencies. Similarly deviations of $\chi'(T,\omega)$ from the isothermal susceptibility are also observed. An inflection point in $\chi''(T,\omega)$ is evident at temperatures corresponding to the temperature of the maximum in $\chi'(T,\omega)$ for each frequency. Both of these features move to higher temperatures with increasing frequency.

ANALYSIS AND DISCUSSION

For temperatures well above T_g, in the paramagnetic regime, the frequency dependent susceptibility is expected to vary according to the Debye relation[19]

$$\chi(\omega)=\chi(\infty)+\frac{\chi(0)-\chi(\infty)}{1+i\omega\tau}, \tag{1}$$

where τ is a unique spin relaxation time and $\chi(0)$ is the isothermal susceptibility in the limit $\omega\tau\ll 1$. $\chi(\infty)$ is the adiabatic susceptibility defined in the limit $\omega\tau\gg 1$. $\chi(\infty)$ is expected to be very small and is generally neglected. This relationship is clearly too simplistic to describe the frequency-dependent susceptibility of the present samples. Our results indicate the presence of a broad distribution of relaxation times. This can readily be demonstrated within the framework of the analysis introduced by Lundgren *et al.*,[20] in which the Debye relation is extended to account for such a distribution. Assuming that the distribution of relaxation times, $g\{\ln(\tau)\}$ varies slowly with $\ln(\tau)$, Lundgren *et al.* demonstrated a simple relationship between $\chi'(\omega)$ and $\chi''(\omega)$ whereby

$$\chi''(\omega)=\frac{\pi}{2}\frac{d\chi'(\omega)}{d\ln\omega}. \tag{2}$$

The insets to Figs. 3(a) and 3(b) show that this relationship is precisely obeyed for the Dy_7Ni_3 and $Dy_3Y_4Ni_3$ samples over most of the temperature regime. Similar agreement with the Lundgren relation is achieved for all $Dy_xY_{7-x}Ni_3$ samples and all measuring frequencies. Equation (2) has previously been used successfully in the analysis of the frequency-dependent susceptibilities of spin glasses, but to our knowledge this is the first time the relation has been demonstrated in strong RMA systems. For the weaker RMA system, $Dy_xGd_{1-x}Ni$,[21] agreement with the Lundgren relation could be reached only after inclusion of a floating scaling parameter.

The validity of Lundgren's relation for the amorphous $Dy_xY_{7-x}Ni_3$ alloys indicates that $g\{\ln(\tau)\}$ is indeed very broad and slowly varying with $\ln(\tau)$ at low temperatures. However, as the temperature increases beyond T_g a discrepancy between the data and the Lundgren relation develops. It is likely that this slight discrepancy at higher temperatures results from a narrowing of the distribution of relaxation times; it can no longer be safely assumed that $g\{\ln(\tau)\}$ varies slowly with $\ln(\tau)$. At still higher temperatures, in the paramagnetic regime well above T_g, $g\{\ln(\tau)\}$ is best described as a δ function appropriate to a single spin relaxation time τ_f.

We believe that the evolution of $g\{\ln(\tau)\}$ with decreasing temperature is a consequence of the growth of Dy spin correlations, or clusters of Dy spins, within the amorphous paramagnetic $Dy_xY_{7-x}Ni_3$ matrix. It is likely that such clusters are related to the short range spin correlations observed in our small angle neutron scattering measurements on the

same alloys. However, we cannot yet establish whether these clusters are rigidly correlated entities, or are essentially dynamic in character.

We have found that it is possible to account for the frequency and temperature dependence of the complex ac susceptibility of the $Dy_xY_{7-x}Ni_3$ alloys using a simple phenomenological model[22] in which the susceptibility is represented by two contributions. The first contribution represents the susceptibility from those independent spins which are essentially free and for which the Debye equation provides an adequate description. For this contribution we can therefore use Eq. (1) with $\tau=\tau_f$ and $\chi(0)=N_f\chi_f(0)$, where N_f is the fraction of nearly free spins in the sample and $\chi_f(0)$ is their isothermal susceptibility. The adiabatic susceptibility, $\chi_f(\infty)$, of the free spins is neglected and τ_f is assumed to be temperature independent.

The second contribution to the susceptibility is assumed to arise from strongly correlated or clustered spins. For these clustered spins the Debye relation can be suitably modified to account for the resulting distribution of relaxation times $g\{\ln(\tau)\}$, i.e.,

$$\chi(\omega)=\chi(0)\int_{-\infty}^{\infty}\frac{g\{\ln(\tau)\}}{1+i\omega\tau}\,d(\ln\,\tau). \tag{3}$$

For a fraction N_c of clustered spins, the complex susceptibility associated with the second component may be expressed, within the framework of the widely used phenomenological Cole-Cole model of relaxation,[23] as

$$\chi(\omega)=\frac{N_c\chi_c(0)}{1+(i\omega\tau_c)^{1-h}}, \tag{4}$$

where $\chi_c(0)$ is the isothermal susceptibility and τ_c is the average relaxation time of the clusters respectively. Combining Eqs. (3) and (4) provides an expression for the distribution of cluster relaxation times $g\{\ln(\tau)\}$

$$g\{\ln(\tau)\}=\frac{1}{2\pi}\,\frac{\sin(h\pi)}{\cosh[(1-h)\ln(\tau/\tau_c)]-\cos(h\pi)}. \tag{5}$$

This distribution function is almost Gaussian in $\ln(\tau)$ and is centered on $\ln\,\tau_c$. The parameter h determines the width of the distribution and can take any value between 0 and 1. These two extreme values of h represent a δ function and an infinitely wide spread of relaxation times respectively.

The relaxation time of the free spins in metal systems is of the order of 10^{-11} s and, therefore, as $\omega_{\max}=35\,625$ rad/s in the current experiment, we can assume that $\omega\tau_f\ll 1$. Within this limit the Debye equation shows that the free spins do not contribute to χ'' but do contribute an amount $N_f\chi_f(0)$ to χ'. In the following analysis we shall let $N_f\chi_f(0)=A$ and $N_c\chi_c(0)=B$. Separating the complex susceptibility of Eq. (4) into its two components, χ' and χ'', and including the free-spin contribution gives a total susceptibility of

$$\chi'(\omega)=A+\frac{B}{2}\left(1-\frac{\sinh(1-h)p}{\cosh(1-h)p-\sin\,\frac{1}{2}h\pi}\right), \tag{6}$$

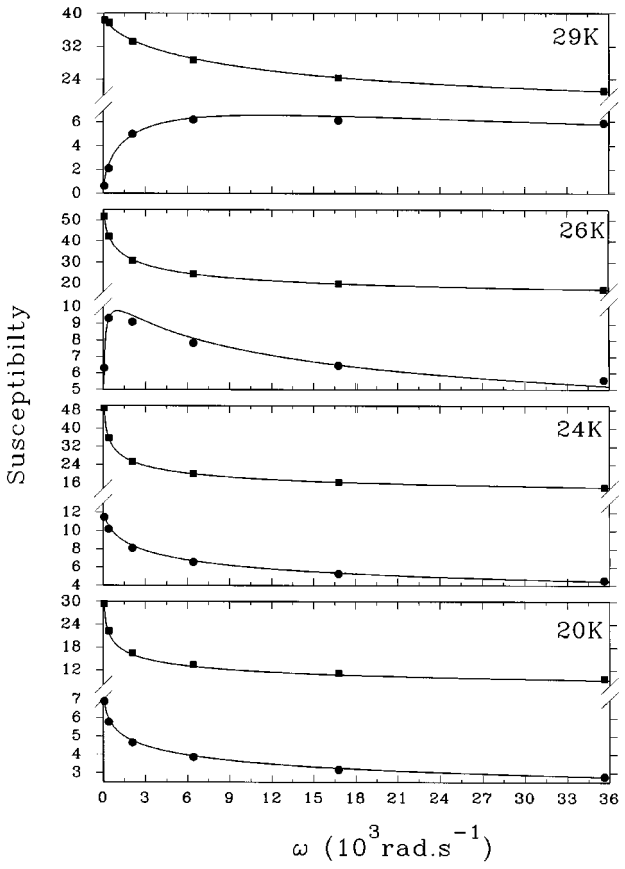

FIG. 4. The real susceptibility (■) and the imaginary susceptibility (●) versus frequency for the $Dy_5Y_2Ni_3$ sample at temperatures of 29, 26, 24, and 20 K. The lines through the data represent a simultaneous fit of Eqs. (6) and (7) to $\chi'(\omega)$ and $\chi''(\omega)$ respectively.

$$\chi''(\omega)=\frac{B}{2}\left(\frac{\cos\frac{1}{2}h\pi}{\cosh(1-h)p+\sin\,\frac{1}{2}h\pi}\right), \tag{7}$$

where $p=\ln(\omega\tau_c)$.

Equations (6) and (7) have been fitted simultaneously to the experimental values of $\chi'(\omega)$ and $\chi''(\omega)$ at discrete temperatures to obtain the four independent parameters A, B, h, and τ_c as functions of temperature. An example of the fitted data is presented in Fig. 4 which shows both $\chi'(\omega)$ and $\chi''(\omega)$ for the $Dy_5Y_2Ni_3$ sample at four different temperatures. Equations (6) and (7) are found to model all the data over the entire temperature and frequency range covered in this experiment.

Figures 5(a) and 5(b) show the temperature dependence of the parameter A (the free-spin susceptibility) and the total isothermal susceptibility $\chi_T(0)$ for the Dy_7Ni_3 and $Dy_3Y_4Ni_3$ samples respectively. $\chi_T(0)$ is the sum of the isothermal free spin (A) and clustered spin (B) contributions to the susceptibility. This parameter is presented in order to allow a direct comparison between the predicted total isothermal and experimental susceptibilities. From Figs. 5(a) and 5(b) it is evident that, even at the relatively low frequency of 11 Hz, the measured low temperature susceptibility is very different to the fitted isothermal, or ''dc'', susceptibility $\chi_T(0)$, although the two do converge at higher temperatures.

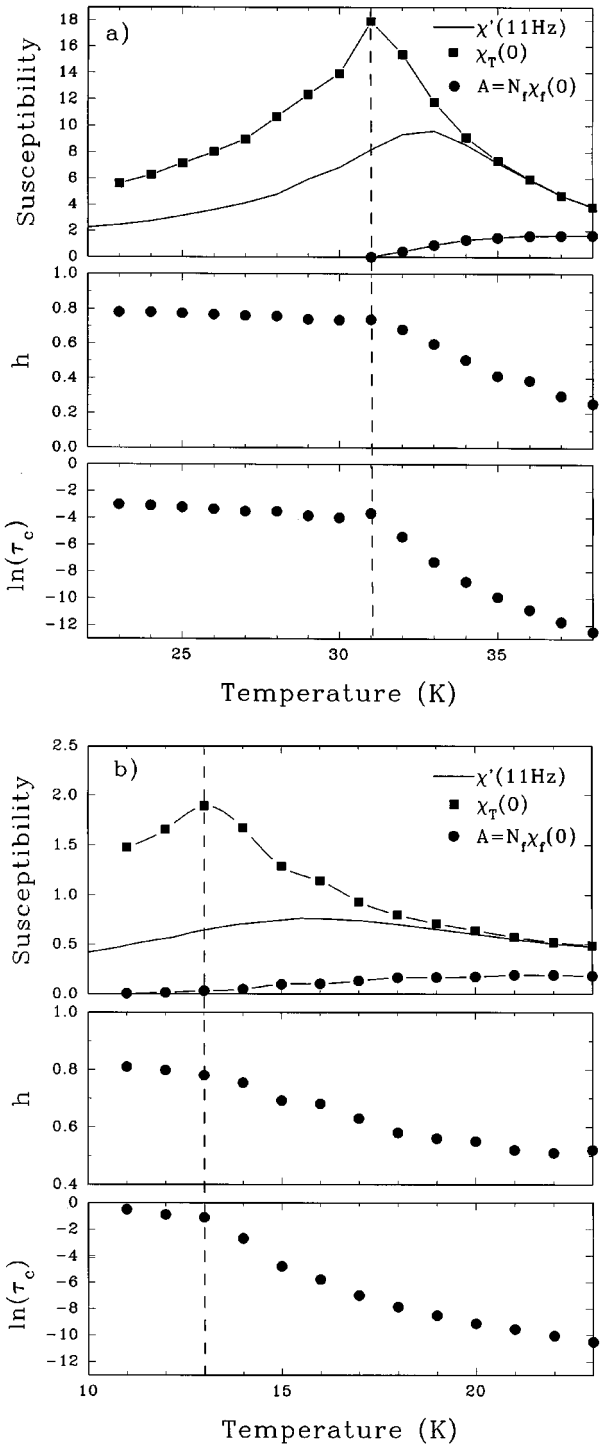

FIG. 5. Temperature dependence of the parameters obtained from a simultaneous fit of Eqs. (6) and (7) to $\chi'(\omega)$ and $\chi''(\omega)$ for (a) Dy_7Ni_3 and (b) $Dy_3Y_4Ni_3$. A is the isothermal free spin susceptibility; $\chi_T(0)$ is the total isothermal, free plus clustered spin, susceptibility; h is the width of distribution of relaxation times; and τ_c is the average cluster relaxation time.

Interestingly the fitted parameters do not suggest a continuous Curie-like increase in $\chi_T(0)$ with decreasing temperature as might be expected for superparamagnetic spin clusters. Instead a peak in $\chi_T(0)$ at a finite temperature, $T_g(0)$, is observed. While the maximum in $\chi_T(0)$ occurs because those

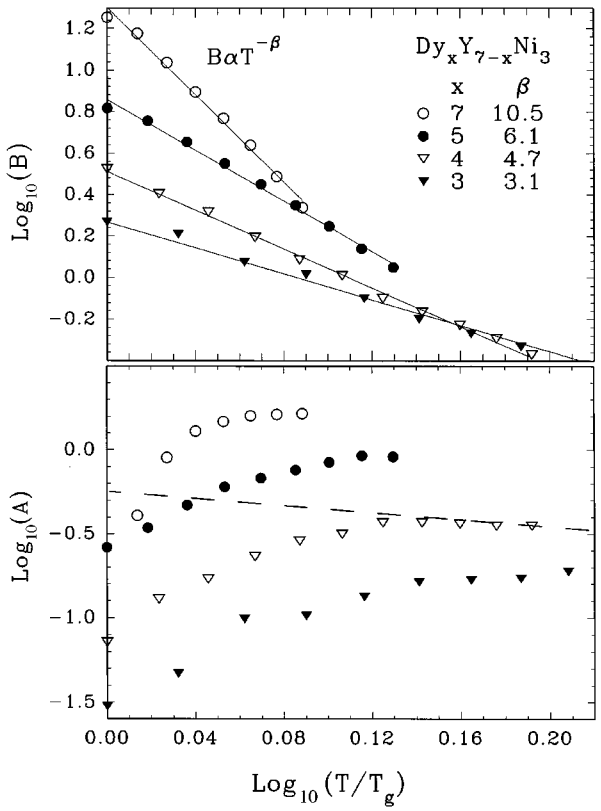

FIG. 6. The parameters A (the isothermal free spin susceptibility) and B (the isothermal clustered spin susceptibility) as functions of the reduced temperature T/T_g for the $Dy_xY_{7-x}Ni_3$ series. The dashed line indicates the slope expected for a simple, Curie-like, $1/T$-dependent susceptibility.

clusters with extremely long relaxation times no longer contribute to the susceptibility, i.e., they are effectively blocked on all time scales, the abrupt change in $\chi_T(0)$ at $T_g(0)$ does suggest that there may be a true magnetic transition at $T_g(0)$. Moreover, with increasing Dy concentration the maximum in $\chi_T(0)$ becomes significantly sharper indicating a developing cooperative behavior between clusters.

A constant, i.e., temperature independent, number of free and clustered spins might be expected to result in a simple $1/T$-like dependence of the susceptibility above $T_g(0)$. Figure 6 shows this is not the case. Instead, the clustered spin contribution to the susceptibility, B, has a temperature dependence of the form $B \propto T^{-\beta}$ where β=3.1, 4.7, 6.1, and 10.5 for the x=3, 4, 5, and 7 samples respectively. Cluster growth is apparently more rapid at higher Dy concentrations. In contrast A decreases with temperature, although not according to any simple power law. This indicates that, on approaching $T_g(0)$, the marked increase in the number of clustered or correlated spins is at the expense of the number of free spins, as might be expected. Below $T_g(0)$, the number of free spins is very small, as evidenced by the extremely small value of A for all samples.

The rapid growth of clusters as $T_g(0)$ is approached from above is accompanied by a large increase in both the width of the relaxation distribution, h, and the average cluster relaxation time, τ_c, as can be seen in Figs. 5(a) and 5(b) for the Dy_7Ni_3 and $Dy_3Y_4Ni_3$ samples. The rapid increase of h and τ_c is curtailed rather abruptly at $T_g(0)$ with both parameters

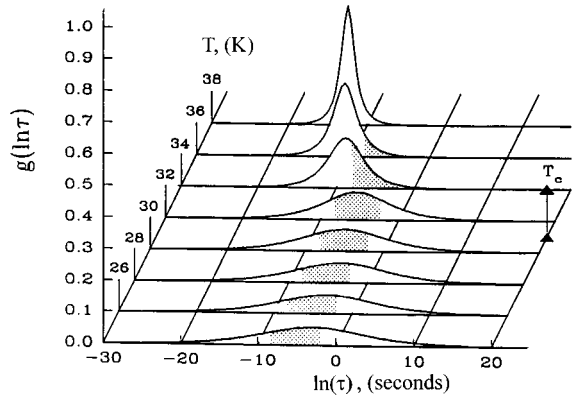

FIG. 7. The evolution with temperature of the distribution of relaxation times, $g\{\ln(\tau)\}$, for Dy_7Ni_3. The shaded regions show the effective time window of the present ac susceptibility measurements.

increasing only slightly in the temperature range below $T_g(0)$. This indicates limited cluster growth below $T_g(0)$ in full accord with the results of small angle neutron scattering experiments[17] which show that the magnetic correlation length in these samples increases with decreasing temperature down to T_g whereupon it remains constant.

In Fig. 7 we present the evolution of the distribution function of relaxation times $g\{\ln(\tau)\}$ for Dy_7Ni_3 obtained via substitution of the parameters h and τ_c into Eq. (5). The effective time window of the experiment, represented by the shaded regions in Fig. 7, is very close to the center of the distribution of relaxation times. This is the reason for the extremely pronounced frequency dependent susceptibility observed for the $Dy_xY_{7-x}Ni_3$ samples. Below $T_g(0)$ the distribution function is so flattened that a large proportion of the magnetic clusters have relaxation times well in excess of one second.

CONCLUSIONS

In this paper we have presented complex, frequency dependent ac susceptibility measurements on amorphous $Dy_xY_{7-x}Ni_3$, a model strong RMA system. For all concentrations the approach to the speromagnetic phase with decreasing temperature is characterized by the growth of magnetically correlated Dy spins, or clusters, which evolve from the reservoir of free Dy spins. Correspondingly, the width of the distribution of magnetic relaxation times $g\{\ln(\tau)\}$ broadens considerably with decreasing temperature, while the mean of this distribution increases. At a characteristic, concentration-dependent temperature, $T_g(0)$, cluster growth and the evolution of the spin relaxation spectrum is curtailed. In this respect the present measurements of the dynamic spin correlations complement our earlier small angle neutron scattering measurements of the static spin correlations in the same $Dy_xY_{7-x}Ni_3$ alloys.[17] It is likely that further growth of the spin correlations below $T_g(0)$ is inhibited by the strong random anisotropy, which, according to theory limits the magnetic correlation length, ξ, to $\xi \propto (J/D)^2$.

The peak in the estimated total isothermal susceptibility, $\chi(T,0)$, provides some evidence that the transition to the speromagnetic state is cooperative. However, any critical scaling analysis must be performed with considerable caution: the relaxation times determined by the present measurements are so long that equilibrium measurements, even by so-called dc techniques, are precluded.

Finally, the salient feature of the present dynamic susceptibility measurements on amorphous $Dy_xY_{7-x}Ni_3$ alloys is the remarkably long characteristic mean relaxation time observed at and above T_g. This relaxation time, typically of the order of seconds, is some four orders of magnitude longer than that generally reported for spin-glass systems, e.g., $Cu_{95}Mn_5$ (Ref. 22) and $(Eu_{0.4}Sr_{0.6})S$ (Ref. 24), at and above their respective glass temperatures. These unusual spin dynamics highlight a fundamental difference between random anisotropy speromagnets and the spin-glass systems with which they are often compared. We have explored these differences still further using zero-field muon-spin relaxation and dc magnetization techniques in comparative studies of amorphous $Dy_xY_{7-x}Ni_3$ and the analogous simple S-state system $Gd_xY_{7-x}Ni_3$. The results of these studies will be presented shortly.

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