PHYSICAL REVIEW B VOLUME 51, NUMBER 21 1 JUNE 1995-I

# Dynamical scaling in ferric oxide spin glasses

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(Received 6 December 1994)

A stochastic relaxation model for the Mössbauer spectra of ferric oxide spin glasses was used to analyze the spectra for the mixed spinel $Mg_{1+t}Fe_{2-2t}Ti_tO_4$ with composition $t=0.70$. The results compare favorably with previously published results on the system $BaSn_xTi_{2-x}Fe_4O_{11}$ with compositions $x=0.40$ and $x=0.80$, and suggest empirical scaling laws for the spin-order parameter defined as $q=\langle S_z\rangle/S$ and the spin correlation time $\tau_c$ in these ferric oxide spin glasses. It was found that the quantity $\tau_c T_G$ versus $T/T_G$ follows a scaling curve with approximately a power-law dependence below the glass temperature. The order parameter follows a scaling curve $q=1-(T/T_G)^{\beta}$, with a value $\beta=2.48\pm0.19$.

## I. INTRODUCTION

A stochastic relaxation model has previously been used to interpret the $^{57}$Fe Mössbauer effect spectra of the ferric oxide spin-glass system $BaSn_xTi_{2-x}Fe_4O_{11}$, as an alternative to other analyses of spin-glass spectra in terms of static hyperfine field distributions.[1] It was found that the temperature dependence of the complex spectra could be reduced to two physically significant quantities: an order parameter defined as $q=\langle S_z\rangle/S$ and a spin correlation time $\tau_C$. The simple model in Ref. 1 also allowed incorporation of the electric quadrupole interaction to first order in the ferric system, which is generally ignored when applying hyperfine magnetic field distributions. In this work the Mössbauer effect spectra of another ferric oxide spin-glass system $Mg_{1+t}Ti_tFe_{2-2t}O_4$ with $t=0.70$ were analyzed using the spin relaxation model in order to compare with the results of Ref. 1. This system had previously been studied extensively using Mössbauer spectroscopy and magnetization measurements, and the spectra were analyzed in terms of distributions of static hyperfine fields.[2] Thus, this system provides an ideal comparison of the stochastic relaxation model and the hyperfine field distribution method, and apparently constitutes the first system to be analyzed both ways.

Relaxation effects have been considered in the Mössbauer spectra of spinel oxides in the past. Relaxation effects have been reported in the $Zn_xCo_{1-x}Fe_2O_4$ system,[35] using essentially the same type of model as in Ref. 1, except the authors allowed for different relaxation times on the *A* and *B* spinel sites. The authors suggested that relaxation effects should also be observable for other spinel systems, including Mg-Ti ferrites[6] (Ref. 7 gives a good review of work regarding relaxation effects in spinel systems). However, except for Ref. 1, ferric oxide spin-glass systems such as $Mg_{1+t}Fe_{2-2t}Ti_tO_4$ ($t=0.70$) have apparently not been analyzed in terms of relaxation phenomena. We suggest here that relaxation effects in spin glasses as observed in the Mössbauer spectra are important characteristics of these materials.

Of interest in the study of any interacting magnetic system is the scaling behavior of the order parameter with temperature. The definition given above, for example, would be proportional to the magnetization in a (collinear) ferromagnetic system, or proportional to the sublattice magnetization in an antiferromagnet. In the case of a spin glass, this order parameter is local, that is, corresponding to an individual spin, where the spins are essentially "frozen" into random positions in the low-temperature spin-glass "phase." This differs from a paramagnetic material which may well show spin relaxation effects but with zero order parameter. In this work an empirical scaling law for the order parameter in these ferric oxide spin glasses will be considered.

Also of interest in the study of spin glasses is the scaling behavior of the spin correlation time $\tau_C$. It has been found[8] with zero-field muon spin resonance (ZF $\mu$SR) methods and neutron spin-echo measurements on the dilute alloy spin glasses (e.g., CuMn, AgMn, AuFe) that the spin correlation times obey a scaling law such that $\tau_C T_G$ is a function of $T/T_G$. The system $BaSn_xTi_{2-x}Fe_4O_{11}$ from Ref. 1 showed single spin-glass transitions for $x=0.40$ and 0.80, but with spin-glass temperatures of 82 and 77 K, respectively, which are too close to convincingly test for such scaling. The system $Mg_{1+t}Ti_tFe_{2-2t}O_4$ with $t=0.70$ is a very similar system but shows a much lower glass temperature of about 12 K. Thus a comparison of the two systems will allow a test for scaling behavior in the spin correlation times in these ferric oxide systems as has been seen in the dilute magnetic alloys.

## II. RELAXATION MODEL FOR MÖSSBAUER SPECTRA

Reference 1 gives details of the stochastic relaxation model used to analyze the spin-glass spectra for the system $BaSn_xTi_{2-x}Fe_4O_{11}$, and these methods will be reviewed only briefly here. Essentially one applies the stochastic relaxation theory of Anderson[9] and Sack[10] to the case of fluctuating hyperfine magnetic fields on a nucleus. This leads to the Dekker model[11] for the Mössbauer spectra. The model includes a "spin-freezing" molecular field and a perpendicular random field which causes transitions between the six $S_z$ states of the ferric ion. As indicated in Fig. 1 of Ref. 1, these two parameters lead to a wide range of spectral line shapes, including very broad spectra similar to those observed in spin-glass systems. For these quantities one can calculate a spin-order parameter $q$ (essentially a Brillouin function of the molecular field) and a spin correlation time $\tau_C$.

In order to include the effect of the small electric quadrupole interaction, it was assumed that the atomic spins and hence hyperfine magnetic fields were directed randomly with respect to the electric field gradient at a given site, so the overall line shape for a spin-glass system consists of a superposition of subspectra having both positive and negative quadrupole shifts. The magnitude of the quadrupole shifts in the spectra were taken to be consistent with the spectra above the glass temperature. It is important to note that the quadrupole shifts lead to line broadening in the random spin system and are often neglected in applying distributions of static hyperfine fields.

Both the $BaSn_xTi_{2-x}Fe_4O_{11}$ system and the $Mg_{1+t}Ti_tFe_{2-2t}O_4$ system have iron atoms on two distinct crystallographic sites which contribute two subspectra. In both Ref. 1 and the present work theoretical site ratios were assumed, with subspectra having different quadrupole shifts, isomer shifts, and hyperfine magnetic fields. The hyperfine magnetic fields on the two sites were determined from the lowest temperature spectrum available, and were then held fixed for all higher temperature spectra. It was assumed, however, that the relaxation parameters were the same for the two sites, mainly to avoid too many free parameters and ambiguities in the curve fitting. Also differences in the recoil-free fraction between the sites were ignored. Besides the baseline and area of the spectra, only the two relaxation parameters were adjusted to fit the temperature dependence on the spin-glass spectra.

## III. ANALYSIS OF THE $Mg_{1+t}Ti_tFe_{2-2t}O_4$, $t=0.70$ SPECTRA

The spectra for the $Mg_{1+t}Ti_tFe_{2-2t}O_4$, $t=0.70$ system were analyzed using the same model as in Ref. 1. Figure 1 shows the spectra and least-squares fits to the relaxation line shapes. From the 4.2-K spectrum hyperfine fields of 423 and 473 kG were obtained for the $A$ and $B$ sites in the spinel structure, with a site ratio $A/B=1.46$ as in Ref. 2, giving an average field value of 443 kG. On the assumption that relaxation effects become minimal at low enough temperatures (that is, beyond the "window" of relaxation times accessible by the Mössbauer technique), the analysis in terms of static hyperfine field distributions and the relaxation model must agree at $T=0$. To test this, the average field from the hyperfine field distributions of Ref. 2 was extrapolated to $T=0$, resulting in a value of 454 kG, in good agreement with the results of the relaxation model. The standard deviation of the two values from the relaxation model is calculated to be $\sigma_B \approx 25$ kG ($T=0$), in comparison with $\sigma_B \approx 50$ kG at 4.2 K from the hyperfine field distribution from Ref. 2. Since the two methods agree in the limit of zero temperature, we conclude that the hyperfine fields extracted using the relaxation model are acceptable. One might suspect, for example, that considerable ambiguity exists in the determination of the fields using the relaxation model, but these results suggest otherwise. The magnitudes of the hyperfine fields on the two sites were held constant for all higher temperature spectra.

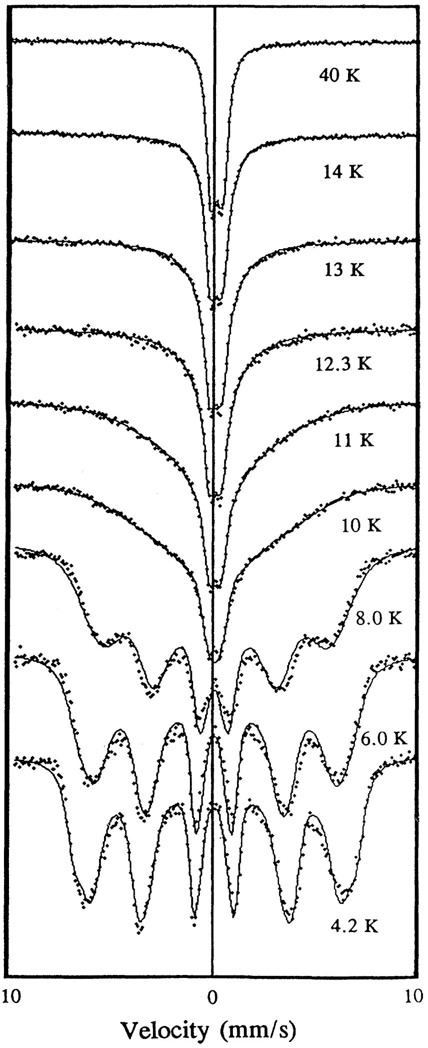

FIG. 1. Mössbauer effect spectra and least-squares fits for the $Mg_{1+t}Ti_tFe_{2-2t}O_4$, $t=0.70$ system.

The electric hyperfine parameters were held constant for the spectra with values consistent with the higher temperature spectra (e.g., $T=40$ K). Thus although the relaxation model considered here does not allow for the *extraction* of the isomer shifts and quadrupole shifts from each spectra, it does allow these effects to be incorporated to first order. Values for hyperfine shifts were $\Delta_I=0.123$ mm/s and $\Delta_Q=0.388$ mm/s for the $A$ sites, and $\Delta_I=0.116$ mm/s and $\Delta_Q=0.147$ mm/s for the $B$ sites (isomer shifts are relative to iron metal).

Figure 2 shows the order parameters for both the $Mg_{1+t}Ti_tFe_{2-2t}O_4$, $t=0.70$ system ($T_G=11.5$ K) along with results for $BaSn_xTi_{2-x}Fe_4O_{11}$ with $x=0.40$ ($T_G=82$ K) and $x=0.80$ ($T_G=77$ K) from Ref. 1. The center solid line indicates the scaling function

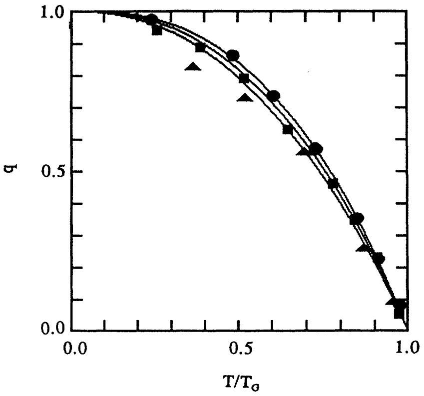

FIG. 2. Order parameter for the ferric oxide spin-glass systems. Circles, $BaSn_xTi_{2-x}Fe_4O_{11}$, $x=0.40$; squares, $BaSn_xTi_{2-x}Fe_4O_{11}$, $x=0.8$; triangles, $Mg_{1+t}Ti_tFe_{2-2t}O_4$, $t=0.70$. The solid curves indicate the scaling function $q=1-(T/T_G)^\beta$ with $\beta\pm\delta\beta=2.48\pm0.19$.

$q=1-(T/T_G)^\beta$ with the best estimate $\beta\pm\delta\beta=2.48\pm0.19$. The outer solid lines indicate similar curves using $\beta+\delta\beta$ and $\beta-\delta\beta$. Note that the scaling function used differs from that used in ferromagnetic systems, where one might take $q=(1-T/T_G)^\beta$ near the Curie temperature . A determination of $\beta$ in this case yielded a value $\beta\pm\delta\beta=0.46\pm0.16$ with a poor fit to the data. It would appear that the scaling law defined above better describes the experimental values, and does so for all temperatures. It is interesting to note that the value determined for the scaling exponent is rather close to the value of $S=\frac{5}{2}$ for the trivalent iron, that is, $\beta\approx S$.

Figure 3 shows a scaling plot of the quantity $\tau_C T_G$ versus $T/T_G$. Again, the two systems considered here obey a similar scaling law. Note that this is a different scaling function than that followed by the dilute magnetic alloys as determined by $\mu$SR in Ref. 8. For the ferric oxide spin glasses one can infer an empirical scaling relation $a=a_0 10^{-\alpha x}$, with $x=T/T_G$, $a=\tau_C T_G$, with values $a_0\approx 2\times10^{-5}$ s K and $\alpha\approx2$, for $T<T_G$. The data are consistent with ZF $\mu$SR measurements obtained for the system 0.9($FeTiO_3$)-0.1($Fe_2O_3$) above the glass temperature,[12] although this system has both $Fe^{2+}$ and $Fe^{3+}$ species, unlike the purely ferric oxides considered here. In Ref. 12, the data were fitted with Fulcher's law, which has a divergence of the spin correlation time at the glass point, but the results in this work suggest that such a divergence does not exist, despite a great slowing of the spin fluctuations as the system passes through the glass temperature on cooling. Apparently it is still an open question whether spin correlation times diverge below the glass temperature or just become very long and finite.[13] Work is currently underway to explain the temperature dependence of the spin correlation time in these materials.

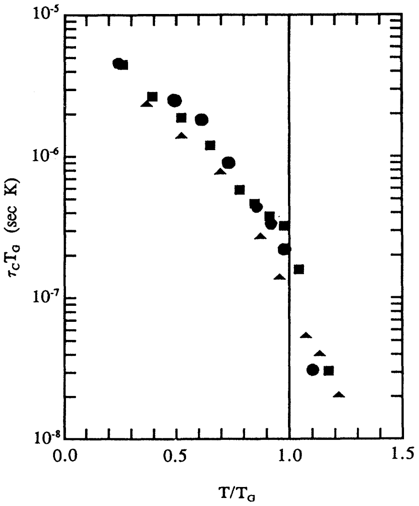

FIG. 3. Scaling plot of the spin correlation time $\tau_C$ for the ferric oxide spin glasses. Circles, $BaSn_xTi_{2-x}Fe_4O_{11}$, $x=0.40$; squares, $BaSn_xTi_{2-x}Fe_4O_{11}$, $x=0.8$; triangles, $Mg_{1+t}Ti_tFe_{2-2t}O_4$, $t=0.70$.

## IV. CONCLUSION

An analysis of the spin-glass system $Mg_{1+t}Ti_tFe_{2-2t}O_4$, $t=0.70$ along with previous analyses of the system $BaSn_xTi_{2-x}Fe_4O_{11}$ with $x=0.40$ and 0.80 suggest scaling behavior of the order parameter and spin correlation times which may hold for all ferric oxide spin glasses. For a scaling function $q=1-(T/T_G)^\beta$ a value for the scaling exponent $\beta=2.48\pm0.19$ was determined. The spin correlation times for these ferric oxides scale in a similar fashion to those in dilute magnetic alloy spin glasses, with the quantity $\tau_C T_G$ versus $T/T_G$ obeying a "universal" scaling function. This scaling function for the ferric oxides is not the same as that for the dilute magnetic alloy spin-glass systems, but nonetheless suggests that such scaling behavior may be a general property of spin glasses. The occurrence of this scaling behavior in both the order parameter and the spin correlation time also suggests that an interpretation of the spectra in terms of relaxation behavior is appropriate.

Further analysis of the Mössbauer effect spectra of the ferric oxide systems would provide more evidence for scaling behavior. In particular, ZF $\mu$SR results on the same systems discussed here would provide the best evidence for a spin relaxation model for the Mössbauer spectra. One might also consider a *spectrum* of relaxation times to characterize the spin-glass state, where the times presented here may represent an average (Ref. 13 gives an excellent review of experimental work on relaxation in spin glasses, including a summary of ZF $\mu$SR results). Such an analysis of the Mössbauer spectra would probably present similar problems as is seen in extracting distributions of hyperfine fields from the spectra, such as pathological degeneracy of the fitting parameters On the other hand, analysis of the spectra for dilute magnetic alloys such as AuFe in terms of a similar relaxation model would provide a comparison of the spin correlation times determined by the present method and ZF $\mu$SR. Such a model would be more complicated due to the presence of "true" hyperfine field distributions in the alloys, but is currently being undertaken, as well as reanalysis of the $Mg_{1+t}Ti_tFe_{2-2t}O_4$ system for other compositions $t$, which shows reentrant ferromagnetic–spin-glass behavior.

## ACKNOWLEDGMENT

We are indebted to R. A. Brandt for kindly providing the spectral data for the $Mg_{1+t}Ti_tFe_{2-2t}O_4$ system some time ago.

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