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Spin freezing of the spin-glass $Fe_{0.25}Zn_{0.75}F_2$: a Mössbauer study

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Abstract We report ^{57}Fe Mõssbauer measurements in the diluted Ising antiferromagnet $Fe_{0.25}Zn_{0.75}F_2$ at temperatures between **4.2** and 28 K. DC susceptibility measurements in the same sample show a spin-glass phase at a freezing temperature Tg = 10K. We found that for this concentration there is a competitive coexistence of spin-glass behaviour and antiferromagnetic order. The freezing temperature is frequency-dependent and follows a power law.

The phase transition behaviour of random magnetic systems continues to attract a great deal of attention both theoretically and experimentally. Very recently it has been shown that an antiferromagnet with intra-sublattice frustration shows a rich (H-T) phase diagram[1] (H=magnetic field, T=temperature). For H=0, a coexistence of antiferromagnetic (AF) and spin glass (SG) phases is observed. As H increases the SG phase is dominant. It has been shown recently[2] that the diluted Ising AF $Fe_xZn_{1-x}F_2$ with $x \leq$ **0.31** displays many features of a three-dimensional short-ranged SG system. At high Fe concentrations ($x > 0.4$), under a uniform externa1 magnetic field, this system is recognized as an excelent experimental realization of a $d = 3$ random-field Ising model(RFIM) system[3]. DC susceptibility measurements for samples with concentrations x $\leq$ **0.25** reveal a **SG** phase appearing at low temperatures[2]. In particular for x = 0.25, the irreversibility temperature is at Tg = 10K. For this same sample ($x = 0.25$) AC-susceptibility measurements confirm the features of SG behaviour: it has a peak in T whose amplitude decreases and which shifts to higher T as frequencies increase; as H_0, an externl applied field parallel to the AC field, increases, the peak is reduced but does not shift in T [4]. By Mössbauer spectroscopy measurements in the same sample we have observed that a competitive coexistence of a SG phase an AF order occurs below 21K [5].

In this paper, we report more detailed low-temperature Mossbauer measurements on this system with $x = 0.25$, and an analysis of the frequency dependence of the freezing temperature.

Figure 1 shows the Mõssbauer spectra at 28, 21, 12 and 4.2 K. At 21 K we observe a superposition of a magnetic hyperfine spectra attributed to an SG phase and a doublet with broad linewidths which correspond to short-range AF order, as was shown previously [5]. Above 28 K we observe a paramagnetic quadrupolar spectrum with narrow linewidts. For T $\leq$ 28K the spectra show an asymetrical doublet with broad linewidth. At 4.2 K the spectrum is similar to that of pure FeF_2, but with a smaller hyperfine magnetic field (H_{hf}) and larger linewidths due to the Zn dilution[6]. This is an indication that the H_{hf} stays perpendicular to the principal axis of the electric field gradient *(EFG)*. For $4.2K \leq$ T $\leq$ 21K the spectra were fitted assuming a superposition of two static and gaussian distributions of hyperfine fields and diagonalizing the complete Hamiltonian. The P(H) distribution is shown in the right part of figure 1. This gaussian behaviour of P(H) is in accordance with Monte Carlo simulations[7] and theoretical results[8]. The introduction of a linear correlation between H_{hf} and the quadrupolar splitting (ΔE_Q) does not improve the fit and we obtain a very small correlation factor. The introduction of the *EFG* asymetry parameter η (value between *0.6* and *0.8)* is necessary in order to obtain good fits.

According to the AC-susceptibility (χ_{ac}) measurements, the cusp temperature depends on the measuring frequencies f [4]. Many researchers have attempted to explain this phenomenon by introducing a time relaxation distribution for the spin systems. As the temperature decreases the relaxation time τ begins to distribute

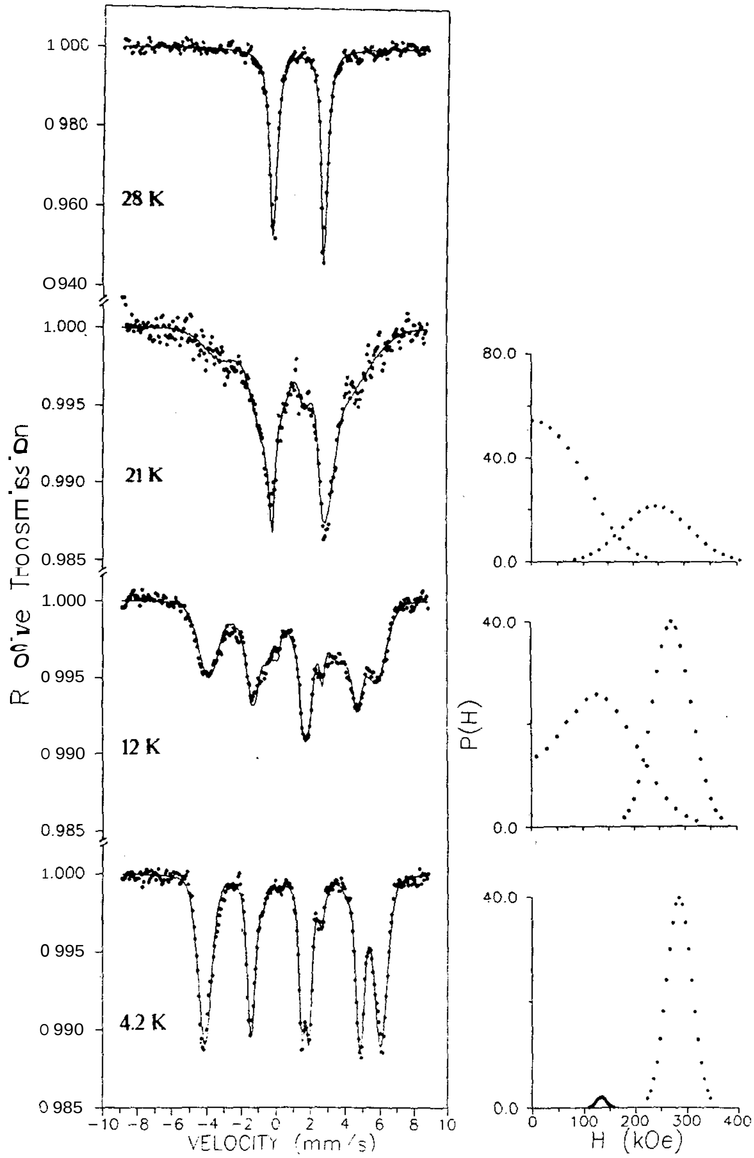

Fig. 1 – ^{57}Fe Mössbauer spectra at 28, 21, 12 and 4,2K in $Fe_{0.25}Zn_{0.75}F_2$. Full lines are fittings with two static, gaussian distributions of H_{hf}. The P(H) curve is shown on the right of the corresponding spectrum.

widely, and the distribution moves toward longer times and spreads broadly. Below the temperature $Tg(f)$ at which a characteristic value r, of the relaxation time becomes equal to f^{-1}, only magnetic moments with $r \leq \tau_c$ contribute to AC-suceptibilities, and χ_{ac} decreases below $Tg(f)$. Thus we can recognize $Tg(f)$ as a temperature where AC-susceptibility shows a maximum. Aruga et al.[9] have shown that the frequency law explaining the dependence of $Tg(f)$ is of the form

$$f = f_0[(Tg(f) - Tg(0)/Tg(f)]^{z\nu} \tag{1}$$

where z is the dinamic exponent and v the critical exponent of the correlation length.

Mõssbauer spectroscopy gives information concerning a single spin time-averaged within the nuclear Larmor precession time τ_L of ^{57}Fe. In the case of $Fe_{0.25}Zn_{0.75}F_2$ τ_L is estimated to be 1.6 x 10^{-7}s, and f $\cong$ 6.1 x 10^6 Hz. By Mõssbauer spectroscopy the transition occurs at $T = 21K$. As the temperature decreases, spin fluctuations slow down gradually. Around 21K, the fluctuation time of some spins becomes comparable to τ_L and the spins appear to freeze. Thus we can consider the temperature of 21K as $Tg(f)$ for f $\cong$ 6.1 x 10^6 Hz. Using the values of $Tg(f)$ obtained by AC-susceptibility[4] and Mõssbauer measurements, we can make a plotting of $Tg(f)$ versus f (Fig. 2). We can see that the power law (1) is obeyed. The values of $z\nu$ and f_0 are around 7 and 9×10^9 Hz, respectivelly. The value of $z\nu$ is in accordance with that obtained by Rezende et.al.[4]. This value is still appropriate for an Ising SG. Nevertheless, the value of f_0 obtained in this work is very diferent from that obtained in $Fe_{0.5}Mn_{0.5}TiO_3$ by Aruga et. al.[9], where $f_0 = 1.4 \times 10^{13}$ Hz. This disagreement can be related to the AF phase present in $Fe_{0.25}Zn_{0.75}F_2$.

Acknowledgments

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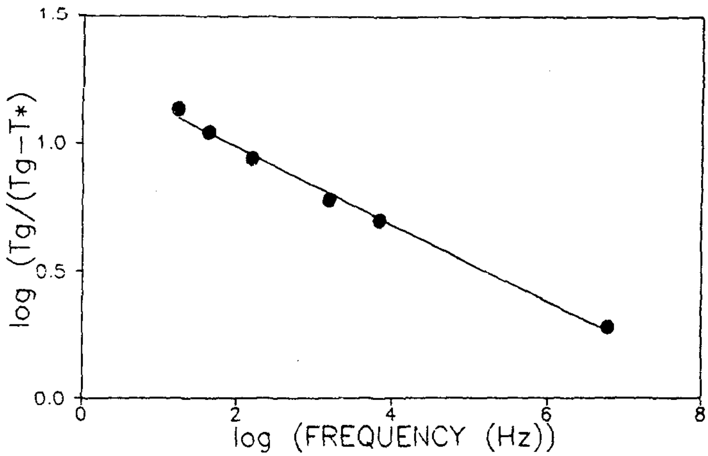

Fig. 2 – Log-Log plot of $Tg(f)/(Tg(f) - Tg(0))$ *versus* frequency.

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Resumo

Apresentamos medidas Móssbauer de ^{57}Fe no antiferromagneto diluido $Fe_{0.25}Zn_{0.75}F_2$ em temperaturas entre 4,2 e 28 K. Medidas de Susceptibilidade magnética DC na mesma amostra mostram a existência de uma fase vidro de spin numa temperatura $T_g = 10$ K. Para esta concentração mostramos que há uma coexistência competitiva entre um comportamento vidro de spin e uma ordem antiferromagnética. A temperatura de congelamento é dependente da frequência e obedece a uma lei de potência.