Revista Brasileira de Física, Vol. 21, nº 2, 1991

Random Field to Spin Glass Crossover Behavior in the Random Magnet $Fe_xZn_{1-x}F_2$

F. C. Montenegro, S. M. Rezende and M. D. Coutinho-Filho
Departamento de Física, Universidade Federal de Pernambuco, 50799, Recife, PE, Brasil

Received August 30, 1900

Abstract An overview of the thermodynamic and critica1 properties of the Ising antiferromagnet (AF) $Fe_xZn_{1-x}F_2$ is presented for a large range of x. For x > 0.4 and under a not very intense field this compound is recognized as a physical realization of the random field Ising model (RFIM) problem. In this regime, a long-range ordered (LRO) AF ground state is established below a critical temperature $T_c(H)$. Both $T_c(H)$ and the equilibrium temperature $T_{eq}(H)$ scale with the universal d = 3 REIM-RFIM crossover exponent $\phi = 1.42$. As x decreases, the LRO AF state becomes unstable for strong magnitudes of the random fields, giving place to a "glassy phase" in the upper part of the (H,T) phase diagram. A striking reversal of the curvature of $T_{eq}(H)$ versus T is observed, now scaling with H as the de Almeida-Thouless replica symmetry breaking line in spin glasses, i.e. $T_N - T_{eq}(H) \approx H^{2/\phi}$ with $\phi = 3.4$. The process evolves to a true Ising spin glass phase dominating the whole (H,T) diagram, for values of x very close an also below the percolation concentration $x_p = 0.24$.

1. Introduction

Essentially all of the experimental work on the random field Ising model' (RFIM) problem has been carried out with frustration-free (anisotropic) diluted antiferromagnets in a uniform field H (DAFF). For small H/J and weak dilution, these systems are equivalent to a ferromagnet with a site-random field[2]. In general, the studies on the RFIM problem are focused on the understanding of the nature of the RFIM ground state and the modifications introduced in the original random exchange Ising model (REIM) critica1 properties when a uniform field is applied collinearly to the easy axis of the DAFFs. Although strong controversies have been generated regarding the deterrnination of the lower critical dimension, d_ℓ, of RFIM

systems, it is now widely accepted that there exists a long-range ordered (LRO) state in presence of weak random fields at d = 3. This is consistent with earlier Imry-Ma[3] energetic arguments about domain formation in RFIM systems, which originally supported $d_\ell = 2$. On the other hand, if the random field is stronger than the field produced by the neighboring spins, then, the spin will follow the random field and no LRO is possible at any temperature. For large H/J ratio, de Almeida and Bruinsma[4], under certain mean-field approximations, have predicted the existence of a glassy phase in strongly diluted AFs of large dimensionality. On the experimental side, however, the magnetic properties of diluted AFs in th strong random field regime have remained virtually unexplored.

In this paper, we present basic thermodynamic and critical properties of the DAFF compound $Fe_xZn_{1-x}F_2$. For $x > 0.4$ and under a not very intense field, this compound is recognized as a physical realization of the (weak) RFIM system. In this regime, a low T AF ordered ground state occurs below a critica1 boundary governed by the REIM-RFIM crossover exponent $\phi = 1.4$. As x decreases, we show that the LRO AF state becomes unstable for strong magnitudes of the random fields, giving place to a "glassy phase" in the upper part of the (H,T) diagram. This situation is acompanied by a profound modification of the critica1 and nonequilibrium behavior of the system. The process evolves to a true Ising spin glass phase dominating the whole (H,T) diagram, for values of x very close and also below the percolation concentration ($x_p = 0.24$).

2. Weak random-field regime

The thermodynamic and critica1 behavior of RFIM systems have been largely studied by severa1 experimental techniques[1]. A crossover has been predicted from REIM to a new RFIM critical behavior, occurring within a crossover region[2]

$$|t| < h_{RF}^{2/\phi}, \tag{1}$$

where

$$t = (T - T_N + bH^2)/T_N \tag{2}$$

is the reduced temperature measured with respect to the mean-field (non-random dilution) phase boundary, $T_N^{MF} = T_N - bH^2$, h_{RF} is the r.m.s. value of the random field and ϕ is the REIM-RFIM crossover exponent.

The reduced mean square value of the random field, for the site-diluted AF, is proportional to the uniform field H according to Cardy's[5] relation

$$h_{RF}^2 = \frac{x(x-1)[T_N^{MF}(1)/T]^2(g\mu_B SH/k_B T)^2}{[1+\Theta^{MF}(x)/T]^2} \qquad (3)$$

In eq. (3), $T_N^{MF}(1)$ is the mean-field Néel temperature in the pure system, and $\Theta^{MF}(x)$ is the (mean-field) Curie-Weiss parameter. From eq. (3), we verify that the magnitude of the random field, for T close to the critical AF-paramagnetic surface [i.e., T $\approx xT_N(1)$], becomes larger as H increases and increases as x decreases. In this section, we will discuss the RFIM properties of $Fe_xZn_{1-x}F_2$, for values of H and x corresponding to weak magnitudes of h_{RF}.

Neutron scattering experiments[1] in several equivalent DAFF compounds have confirmed the existence of a low-T long-range ordered ground state for the (weak) RFIM problem (x > 0.4). With magnetization measurements, we can also identify the equilibrium ground state of a DAFF compound. We have used the so called zero-field cooling (ZFC) and field cooling (FC) procedures to perform field-hysteresis loops, as illustrated in Fig. 1, for a sample of $Fe_xZn_{1-x}F_2$ with x = 0.48. The measurements were made with a vibrating sample magnetometer in magnetic fields up to 6T. The temperatiire was measured with a carbon glass thermometer and controlled to within 1mK in the range 4.2 < T < 60K. The field scan rate was $0.3T/min$ and mean cooling rate $5 - 10K/min$. The field loops made after the sample was cooled in zero field (ZFC) present no detectable hysteresis or time dependence within the stability limits of the AF ground state. These limits are estalished by the (H_{eq}, T_{eq}) points where the field increasing magnetizations differ from the field decreasing ones (see arrow in Fig. 1(c)). The irreversibility line for this sample, shown in Fig. 2, follows the same crossover scaling[1] as the critical temperature $T_c(H)$, i.e., $T_N - T_{eq}(H) = cH^{2/\phi}$, with $4 \approx 1.4$. The irreversibilities which take place when the sample is cooled in a field arise from RF effects, consistent with previous results obtained in this system by other techniques[1]. The

dashed lines in Fig. 1(a)-(c) show the M(H) cycles at several temperatures obtained after cooling the sample in a field H = 6T. In this case, it is known that a large excess magnetization $M = M_{FC} - M_{ZFC}$ develops due to the nucleation of metastable domains structures[6,7]. These studies are consistent with earlier studies in the RFIM systems $Fe_{0.7}Mg_{0.3}Cl_2$[8] and $Fe_{0.47}Zn_{0.53}F_2$[9].

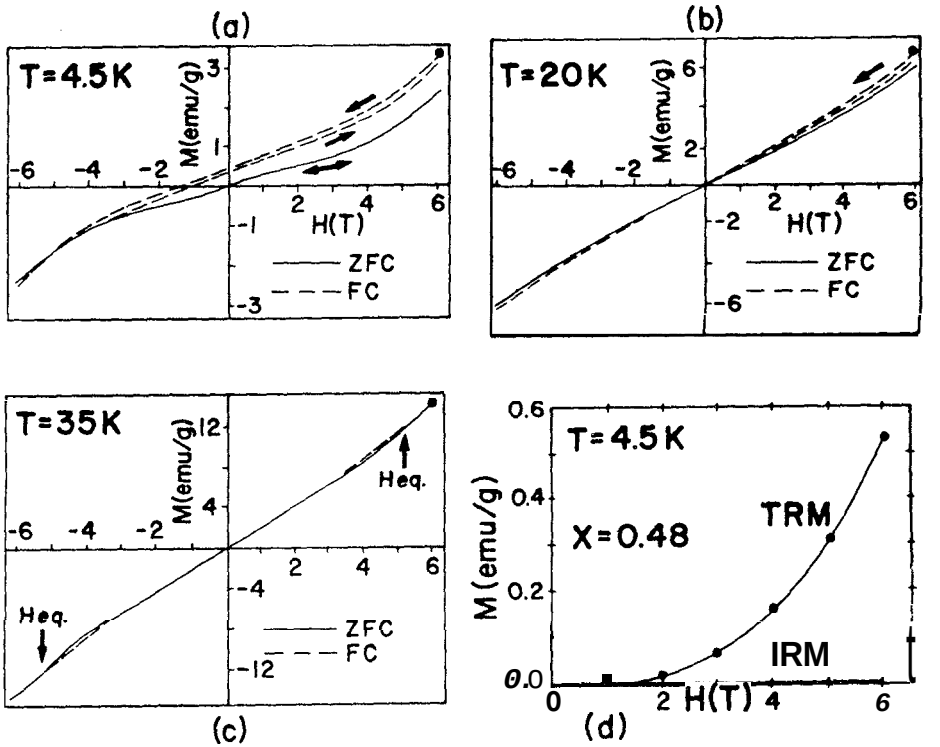

Fig. 1 - M vs H curves following complete field cyclings (as indicated by arrows in (a)) in $Fe_{0.48}Zn_{0.52}F_2$ obtained with ZFC and FC procedures at temperatures $4.5K$ (a), $20K$ (b), and $35K$ (c). The black circles indicate the starting point after cooling the sample. (d) shows TRM vs H and IRM vs H for T = $4.5K$. The full line in TRM is best fitted with a power law, $TRM \alpha H^{\gamma}$, with $\gamma \approx 3.05 \pm 0.05$.

The novel feature of our data for the $x = 0.48$ sample is the asymmetric open cycle, characterizing the FC magnetization curves obtained by reversing the field after it is scanned from $H_0 = 6.0T$ down to zero. It is interesting to note that

at low temperatures M does not vanish when the field is reversed, even with a value as large as $H = -H_0 = -6.0T$. Note also that at $4.5K$ only a small fraction of the remanent magnetization is lost after such a field reversal. This indicates that the domain structure remains frozen in, possibly because of the narrow wall random bond pinning[7–9]. With increasing T, however, the domain walls broaden and the random bond pinning is less effective in keeping the domain structure unhanged, resulting in domain relaxation (see Fig. 1(b)). On further increasing the temperature, the domain structure relaxes completely so that $M_{FC} = M_{ZFC}$, except in the vicinity of the equilibrium phase boundary (Fig. 2) as illustrated in Fig. 1(c).

The H dependences of the isothermal- and thermo-remanent magnetizations (IRM and TRM) are shown in fig. 1(d), for T=4.5K. The IRM is measured by first cooling the sample in ZFC to a given temperature T, then, increasing the field to a value H and, finally, turning it off to measure M. The TRM is measured after turning the field off in a FC cycle. For the $x = 0.48$ sample, IRM=0, in the field range of the experiments ($0 < H < 65$kOe), at all temperatures. The behavior of IRM and the ZFC magnetization entail that the systern develops AF order only if the field is applied after cooling in H–O. On the other hand, when the system is cooled in a field, the irreversible bchavior seen in curves 2(a,b) and the H dependence of TRM (see fig. 2(d)), indicates that the system lies in a domain state. The behavior of the remanent magnetizations in the RFIM regime ($x = 0.48$) can be contrasted to the spin glass case (x = 0.25) shown in fig. (6.d). One question of current debate is whether the random field TRM is associated with the volume[6] or the surface[7] of the frozen in domains. The solid line in fig. 2(d) is a fit to AM α $H_0^{\nu_H}$ with $\nu_H = 3.05$ indicating that in the field range 2.0-6.0T the excess magnetization arises from volume contributions ($\nu_H = 3.0$) [6]. It is not unlikely, however, that at lower fields the surface contribution becomes more important because of the larger domain size, as it has been verified in $Fe_{0.7}Mg_{0.3}Cl_2$[8].

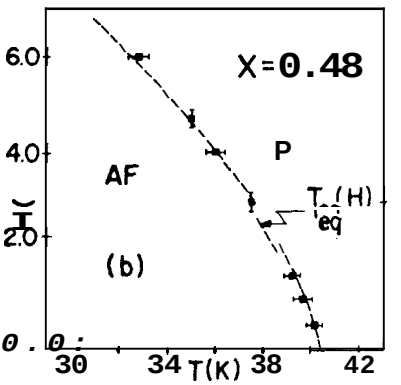

Fig. 2 - Fig. 2 - Irreversibility phase diagram for $Fe_{0.48}Zn_{0.52}F_2$.

3. Crossover from RFIM to spin glass-like behavior

The evolution of the d = 3 weak RFIM behavior to a glassy phase in its strong limit was experimentaly detected and reported for the first time by magnetization measurements[10,11] on the Ising antiferromagnet $Fe_{0.31}Zn_{0.69}F_2$. Later on, Faraday rotation[12] (FR) and neutron scattering[13] techniques have explored details about the modifications introduced in the critical behavior of this sample. In Fig. 3 we show the measured FR angle O, which is proportional[14,15] to the uniform magnetization as a function of temperature T for three fields, H = 0.5, 1.0, and $1.5T$, following both ZFC and FC procedures. The data reproduce the same qualitative features of the earlier magnetization[10] but are more precise and give much better resolution of the irreversibility temperature. A dramatic hysteresis is seen in the FC and ZFC results below a temperature $T_{eq}(H)$, defined, as discussed in section 2, as the temperature above which all measurements give results independent of the field cycling procedure. In the critical region, the temperature derivative $d\Theta/dT$ v T has been demonstrated to be proportional to the magnetic specific heat C, in dilute antiferromagnets[14]. $d\Theta/dT$ vs T is shown in Fig. 4 for H = 0.05, 0.5 and $1.0T$. Similarly to samples with higher concentrations[1] $x > 0.4$, C, exhibits a symmetric peak rounded by the extreme critica1 slowing down that occurs near the phase transition $T_c(H)$. The observation of a symmetric, dynamically rounded divergence, particularly in the ZFC measurements, is indicative of the existence of

$d = 3$ RFIM critical behavior in the $Fe_zZn_{1-z}F_2$ system[1]. $T_c(H)$ and $T_{eq}(H)$ can be accurately determined from the $d\Theta/dT$ versus T data since neither the peak position nor the point at which hyteresis is observed is strongly dependent upon the measurement time as long as H $\leq 1.5T$. That antiferromagnetic long range ordering is associated with the ZFC peak has been verified in neutron scattering experiments[13] on the same crystal.

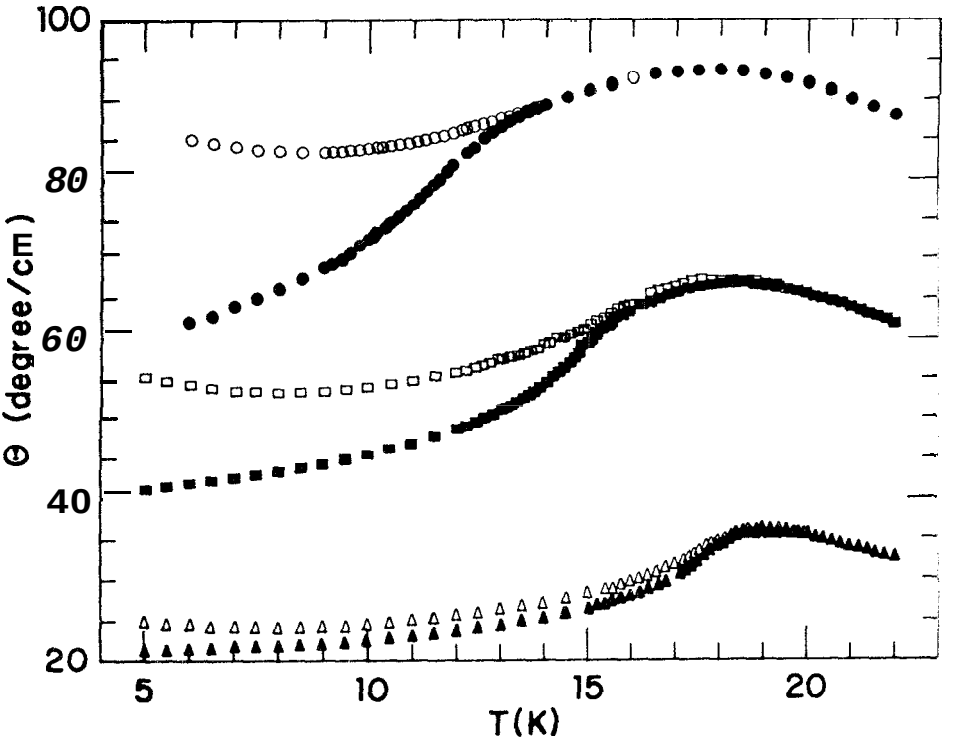

Fig. 3 - 0 vs T for H = 0.5, 1.0,and $1.5T$ for $x = 0.31$. The filled symbols represent FR data taken upon ZFC and the open symbols are for FC. $T_c(H)$ is at the inflection point in Θ versus T of the ZFC data. $T_{eq}(H)$ is the point at which the ZFC and FC data no longer coincide.

Quite different behavior is observed at larger fields. Although Θ vs T piots for $H > 1.5$ appear[12] qualitatively similar to the low field scans shown in Fig. 3, there is, in fact, a crucial difference. While there is an inflection point in Θ versus T for ZFC data, the position and shape of the curve is strongly dependent

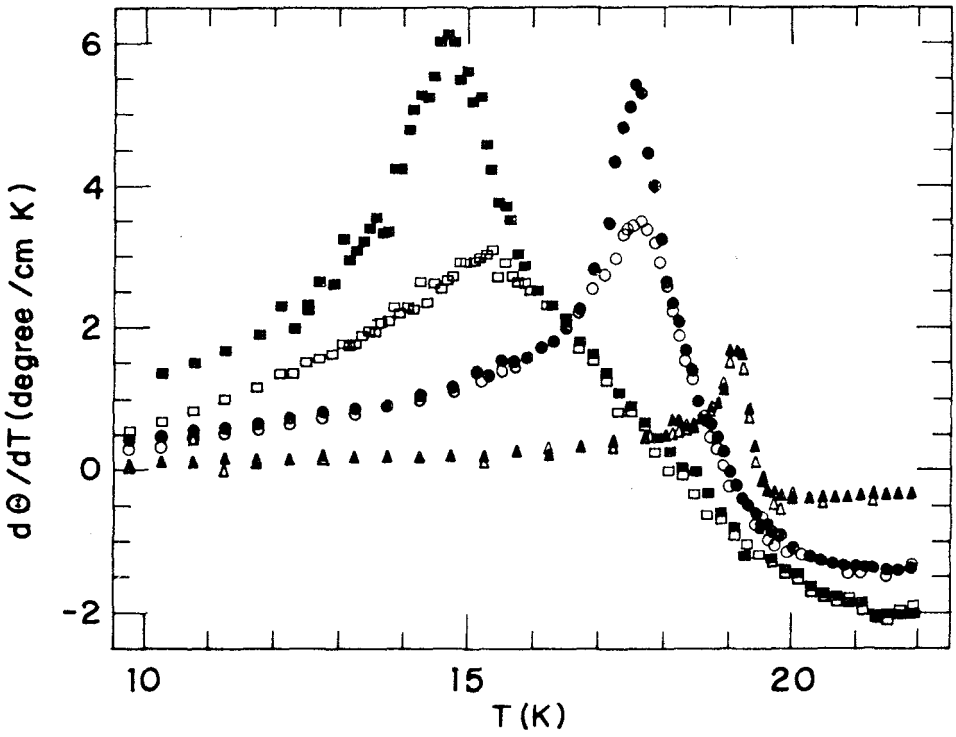

Fig. 4 - $d\Theta/dT$ vs T for H = 0.05, 0.5, and 1.0T for $x = 0.31$. The filled symbols represent data taken upon *ZFC* and the open symbols are for *FC*. The *ZFC* peaks are proportional to the specific heat and are dynamically rounded. The *FC* peaks are suppressed since the long range AF ground state is not achieved via this procedure.

on the scanning rate. Hence, this feature cannot readily be associated with a phase boundary (i.e. $T_c(H)$), as is the case for small fields. Indeed, the neutron scattering measurements[13] do show that the low T region is not associated with AF LRO. The value of $T_{eq}(H)$ obtained, on the other hand, is not significantly rate dependent for the range of rates used in these measurements and could, therefore, be consistently determined.

All the data for $T_c(H)$ and $T_{eq}(H)$ for $0 < H < 7T$ are shown in Fig. 5. The lower field portion of the diagram corresponds to the RFIM behavior observed previously for $x > 0.4$, where the crossover exponent $\phi = 1.4$ is the same as

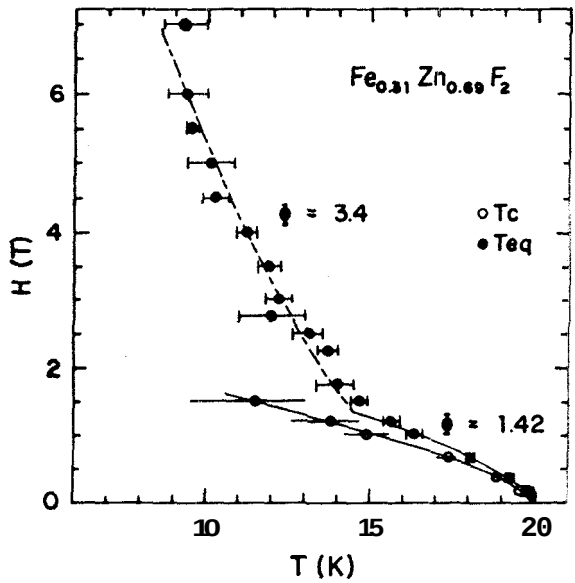

Fig. 5 - The T vs H phase diagram of the $Fe_{0.31}Zn_{0.69}F_2$ RFIM system. Error bars are given for $T_{eq}(H)$ measurements. The horizontal bars through the ZFC $T(H)$ data represent the dynamical rounding of the transition as indicated by the inflection points in $d\Theta/dT$ versus T. The solid and dashed curves represent scaling behavior for low and high field with $\phi = 1.42$ and 3.4 respectively. For large H, following ZFC, inflection points in Θ versus T could be observed, but were very time dependent in shape and location. Hence, they are not identified with $T_c(H)$, as was the case for low H.

that seen for $x > 0.46$. However, the large H behavior is a novel feature. A fit to the scaling behavior $T_N - T_{eq}(H) = H^{2/\phi}$, with the fixed value $\phi = 3.4$, is indicated by the dashed curve. $T_c(H)$ cannot be consistently determined, because of the lack of long-range AF ordering, as detected by neutron experiments[13]. The thermodynamic and scaling behavior at large H values is similar to that observed in conventional spin glass systems. The particular choice of $\phi = 3.4$ for the large H scaling of Fig. 5 is motivated by the observation of a similar scaling to the equilibrium boundary[16,17] in $Fe_{0.5}Zn_{0.75}F_2$, which presents all features[16-18] of a canonical Ising spin glass, in the whole (H,T) diagram. The SG characteristics of the x = 0.25 sample will be discussed in the next section.

A monotonic increase with time has been detected in the uniform magnetization at low T, when the field was increased rapidly to large values ($H > 1.5T$). This corresponds to the observation in the neutron scattering experiments[13] that, under similar conditions, the staggered magnetization decreases with time as H increases. This finding confirms the instability of the LRO AF state for $H > 1.5T$.

4. Spin-glass regime

$Fe_xZn_{1-x}F_2$ close and below the percolation concentration $x_p = 0.24$, presents the standard features of a canonical Ising spin glass[16–18]. The contrast between the typical SG history dependence for a sample with $x = 0.25$, and the corresponding behavior for x = 0.48 (see Fig. 1) is evident in the ZFC field hysteresis cycles shown in Fig.6. $Fe_{0.25}Zn_{0.75}F_2$ has a freezing temperature[17] $T_f = 10K$. For T < T, M is completely reversible only at fields above the de Almeida-Thouless (AT) irreversibility line[17] ($H > 5.6T$ at $T = 4.5K$ as indicated by the arrow in the H axis of Fig. 6(a)). Note that although the hysteresis loop shrinks as $T \rightarrow T_f$ (Fig 6(b)), M remains highly nonlinear above T_f (Fig. 6(c)), before reaching the standard pararnagnetic behavior at about $60K$. The AT line shown in Fig. 7 was determined by measuring the irreversibility temperature, $T_i(H)$, at which the FC and ZFC data depart from each other. Below $T_i(H)$ the ZFC magnetization measured on heating is irreversible and relaxes in time towards the FC values. The FC data,on the other hand, are reversible and show no time-dependence (in a time scale of an hour). Also typical of a SG are the data for the H dependences of the isothermal and thermo-remanent magnetizations (IRM and TRM) shown in Fig. 6(d) for $T = 4.5K$. This behavior of IRM and TRM is to be contrasted with that shown in Fig. 2(d), for the $x = 048$ sample.

The SG nature of the $Fe_{0.25}Zn_{0.75}F_2$ sample was confirmed by other meaurements. The ac susceptibility[18] $\chi'(\omega, T)$ has a peak in temperature which reduces in amplitude and shifts to a higher T as the frequency increases. This peak smears down, but does not shift in T as the applied field increases. A Mössbauer study[19] in this sample shows a competitive coexistence of SG and AF order below 21K. The former dominates at $T \approx 10K$, leading to practically the entire crystal in the

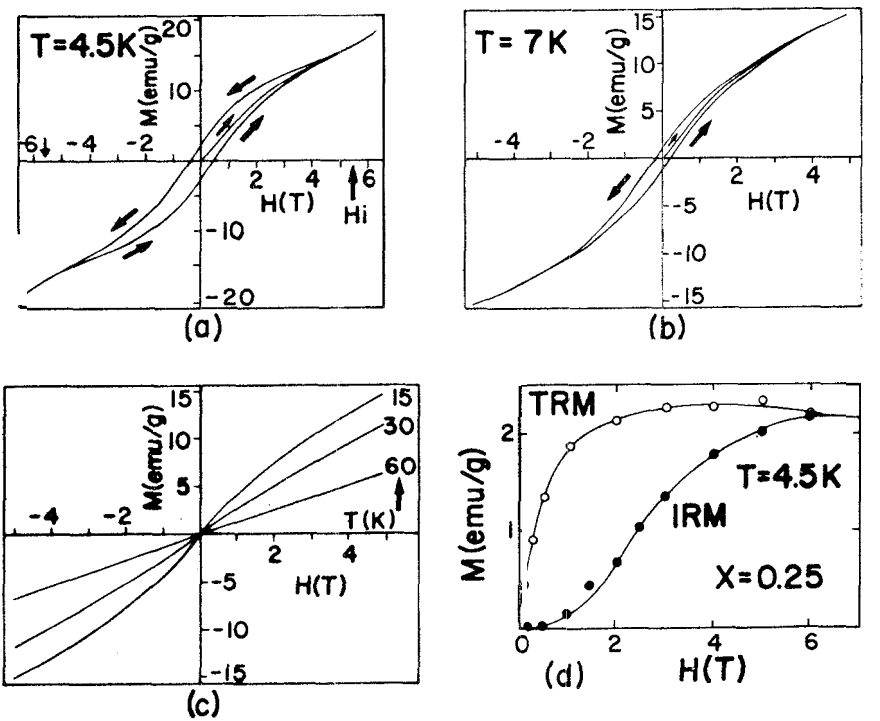

Fig. 6 - M vs H curves for $Fe_{0.25}Zn_{0.75}F_2$ at several temperatures obtained after cooling in H = 0. The field cycling that yields the hysteresis loops is indicated by arrows in (a) and (b). (d) shows the H dependence of IRM and TRM for T = 4.5K.

SG phase. Moreover, static and dynamic scaling analyses of the nonlinear and ac susceptibility[18] data, respectively, yield a consistent set of critica1 exponents in excellent agreement with those found in metallic Ising spin glases. This last finding provides an evidence for uriiversality of Ising spin glasses, a matter of present controversy.

5. Conclusions

From the observations in $Fe_zZn_{1-z}F_2$ at concentrations $x > 0.4$, x = 0.31

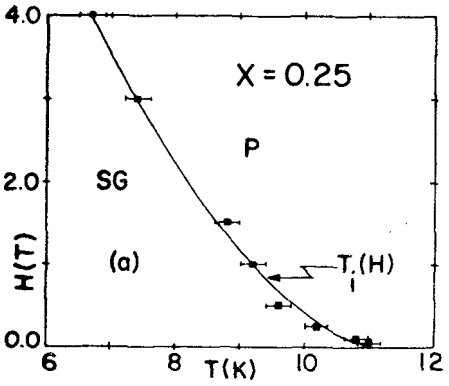

Fig. 7 – Irreversibility phase diagram for $Fe_{0.25}Zn_{0.75}F_2$.

and x = **0.25,** a schematic evolution of the $T_{eq}(H)$ boundary as x approaches x_p is proposed in Fig. 8. The large x regime is entirsly described by scaling with ϕ = **1.42.** Somewhere above x = **0.31,** in addition with the low field behavior with 4 = **1.42,** the high field behavior with ϕ = **3.4,** would first appear. The low field region, corresponding closely to the onset of AF LRO, shrinks rapidly as x approaches x_p and the large H scaling becomes the sole feature of $T_{eq}(H)$. A theoretical understanding of this novel feature of the phase diagram vs x is still a challenging problem.

We have presented an overview of the basic thermodynamic and critica1 properties of the compound $Fe_xZn_{1-x}F_2$ in the whole range of z. In the weak random field regime, the LRO AF ground state is reached by ZFC procedure below a boundary which scales in a universal way in the form $T_N - T_c(H) \approx H^{2/\phi}$ with ϕ = **1.42.** $T_{eq}(H)$ lies just above $T_c(H)$ and scales as $T_N - T_{eq}(H) \approx H^{2/\phi}$, with the same crossover exponent ϕ = 1.42. In an intermediate x region ($x \approx 0.31$), the above scaling behavior for $T_c(H)$ and $T_{eq}(H)$ is found only for low values of H. In the large H region, the behavior is similar to that seen at all H for x = 0.25. $T_{eq}(H)$ scales as $T_N - T_{eq}(H) \approx H^{2/\phi}$ with ϕ = **3.4** as a typical de Almeida-Thouless replica symmetry-breaking line in spin glasses and departs widely from the extrapolation of $T_c(H)$; the large region between them exhibits very slow (SG-like) dynamics and no AF LRO. For x values very close and below the percolation

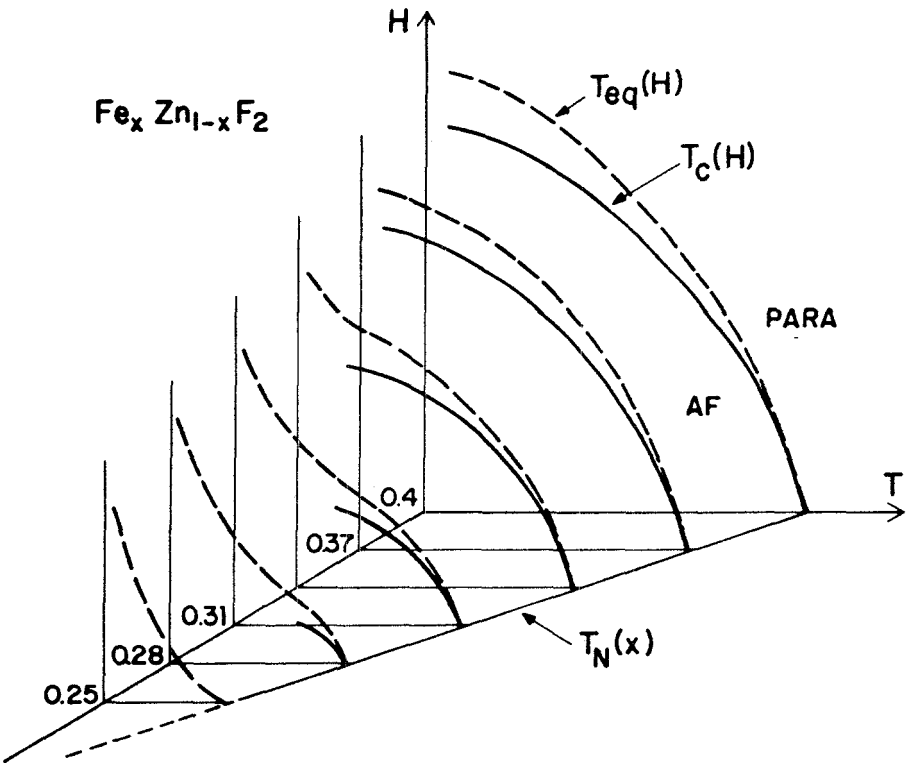

Fig. 8 – The schematic evolution of the H versus T phase diagram for $Fe_xZn_{1-x}F_2$ system with z. As x decreases, the novel high field behavior occurs in $T_{eq}(H)$ probably first above $x \approx 0.37$. As x approaches $x_p = 0.24$, the spin glass portion of the phase diagram increases and the antiferromagnetic portion shrinks. At $x = 0.24$, the antiferromagnetic part has vanished and the only feature remaining is $T_{eq}(H)$, with a de Almeida-Thouless-like curvature, and there is no AF LRO. The small dashed curve represents the x dependence of the crossover from the low to the high H regions.

threshold, the system behaves like a canonical Ising spin glass, in the whole range of H.

We acknowledge U. A. Leitão for collaborating with the magnetization measurements at UFPE, and D. P. Belanger, A. R. King, V. Jaccarino and S-j. Han for helpful discussions and participation in the FR measurements at UCSB and

UCSC. This work was supported by FINEP, CNPq and CAPES (Brazilian Agencies).

References

1. For recent reviews on the RFIM problem, see for instance: V. Jaccarino and A. R. King, Physica A 163, **291 (1990);** D. P. Belanger, Phase Transitions **11, 53 (1988).**
2. S. Fishman and A. Aharony, J. Phys. C 12, **L729 (1979).**
3. **Y.** Imry and S-k. Ma, Phys. Rev. Lett. 35, **1399 (1975).**
4. **J.** R. **L.** de Almeida and R. Bruinsma, Phys Rev. B 37, **7267 (1987).**
5. J. L. Cardy, Phys.Rev. B 29, **505 (1984).**
6. J. Villain, Phys. Rev. Lett. 52, **1543 (1984).**
7. T. Natterman and I. Vilfan, Phys. Rev. Lett. 61, **233** (1988).
8. U. A. Leitão, W. Kleeman, and I. B. Ferreira, Phys. Rev. B 38, **4765** (1988).
9. P. Pollak, W. Kleeman, and D. P. Belanger, Phys. Rev. B 38, **4773 (1988).**
10. F. C. Montenegro, U. A. Leitao, M. D. Coutinho-Filho, and S. M. Rezende, J. Appl. Phys., 67, **5243 (1990).**
11. S. M. Rezende, F. C. Montenegro, U. **A.** Leitão, and M. D. Coutinho-Filho in ***New Trends in Magnetism,*** eds. M.D. Coutinho-Filho and S. M. Rezende, Word Scientific, Singapore **(1990)** p. **44.**
12. F. C. Montenegro, A. R. King, V. Jaccarino, S-j. Han, and D. P. Belanger, Phys. Rev. B, to appear.
13. D. P. Belanger, Wm. Murray Jr., F. C. Montenegro, A. R. King, V. Jaccarino, and R. Erwin, Phys Rev. B, to appear.
14. W. Kleeman, **A.** R. King an V. Jaccarino, Phys. Rev. **B** 34, **479 (1986).**
15. U. **A.** Leitão and W. Kleeman, Phys. Rev. B 35, **8696 (1987).**
16. F. C. Montenegro, S. M. Rezende, and M. D. Coutinho-Filho, J. Appl. Phys. 63, **3755 (1988).**
17. **F.** C. Montenegro, M. D. Coutinho-Filho, and S. M. Rezende, Europhys. Lett. 8, **383, (1989);** see also, F. C. Montenegro, M. D. Coutinho-Filho and S. M. Rezende, J. Physique C8, **1007** (1988)..

18. S. M. Rezende, F. C. Montenegro, M. D. Coutinho-Filho, C. C. Becerra, and A. Paduan-Filho, J. Physique ***C* 8,** 1267 (1988).

19. J. B. M. da Cunha, J. H. de Araújo, L. Amaral, A. Vasquez, J. T. Moro, F. C. Montenegro, S. M. Rezende, and M. D. Coutinho-Filho, Hyperfine Interactions, 54, 489 (1990).

Resumo

Uma abordagem geral das propriedades termodinâmicas e críticas do antiferromagneto (AF) Ising diluído $Fe_xZn_{1-x}F_2$ apresentada para uma larga faixa de valores de z. Para $x > 0.4$ e sob a ação de campos não muito intensos, este composto é reconhecido como uma realização física do modelo Ising com campo aleatório (RFIM). Nesse regime, um estado fundamental com ordenamento de longo alcance (LRO) antiferromagnético é estabelecido abaixo de uma temperatura crítica $T_c(H)$.$T_c(H)$ e a temperatura de equilíbrio $T_{eq}(H)$ escalam com o expoente universal de cruzamento REIM-RFIM $\phi = 1.42$. Quando x decresce, o estado com ordenamento antiferromagnético de longo alcance se torna instável para campos aleatórios fortes, dando lugar a uma "fase vítrea" na parte superior do diagrama de fase (H,T). Uma surpreendente reversão de curvatura em $T_{eq}(H)$ versus T é observada, agora escalando com H como a linha de quebra de simetria de réplicas, de Almeida-Thouless, em vidros de spin, i.e. $T_N - T_{eq}(H) \approx H^{2/\phi}$, com $4 = 3.4$. O processo evolui para uma fase vidro de spin Ising dominando completamente o diagrama (H,T), para valores de x muito próximos e também abaixo da concentração de percolação $x_p = 0.24$.