Influence of frustration on a \(d=3\) diluted antiferromagnet: \(Fe_xZn_{1-x}F_2\)

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Abstract

The influence of a frustrated bond on the magnetic properties of a \(d=3\) uniaxial (Ising) b.c.c. diluted antiferromagnet, with emphasis in the compound \(Fe_xZn_{1-x}F_2\), is investigated by a local mean-field numerical simulation. In particular we find that the initial drop of the saturation staggered magnetization (\(M_S\)) with concentration follows a percolation-like phenomenon characterized by an exponent \(\beta_p\). For the frustrated samples, however, this regime is followed by a second one identified by a “long tail” effect such that \(M_S\) is zero only at the percolation threshold. Our numerical data also confirms a spin-glass phase near this threshold.

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Diluted antiferromagnets (DAFs) have been largely used as testing ground for theoretical models of random magnets[1]. In spite of this, a complete understanding of the microscopic mechanisms behind the rich variety of phases found in these systems is still far from being accomplished. The particular nature of the exchange interactions, the external applied field, as well as the spatial anisotropy give rise to distinct magnetic properties in real systems. With respect to the above parameters random systems with uniaxial anisotropy and short-range interactions can be classified by Ising models belonging to different universality classes. In three dimensions (d=3) the insulating DAF compound $Fe_xZn_{1-x}F_2$, with a centred tetragonal lattice, is one of the most studied experimental realizations of such models[1-4]. Due to a strong single-ion anisotropy, pure $FeF_2$ is considered an archetypal Ising system. The partial random substitution of $Fe^{+2}$ by $Zn^{+2}$ ions in this compound make it to behave magnetically as a random-exchange Ising model (REIM). The application of a uniform field parallel to its easy direction causes a crossover from REIM to the random-field Ising model (RFIM) critical behavior[1,5]. For highly diluted samples, random fields of strong magnitudes make the AF long-range order (LRO) unstable and nucleates a “glassy” phase in the upper part of the $(H, T)$ phase diagram[6]. In the vicinity of the percolation threshold ($x_p \approx 0.24$) a true Ising spin glass (SG) phase was found to occur below a freezing temperature in a sample of $Fe_{0.25}Zn_{0.75}F_2$ [4]. For the appearance of a SG behavior randomness and frustration are considered essential ingredients. However, the effects of very weak frustrated interactions, eventually present in short-ranged random magnets, have been a priori neglected as compared with the ones caused by stronger interactions[2]. The opposite limit, i.e., systems with weak disorder and strong frustrated interactions are also of considerable interest.

In a previous paper[3], we presented results of a zero-field $(H = 0)$ numerical
simulation which showed the dramatic role played by a weak frustrated interaction in the antiferromagnetic \((M_S)\) and spin-glass \((Q)\) order parameters of the \(Fe_xZn_{1-x}F_2\), leading the system to the SG phase observed\([4]\) in the strong-dilution regime \((x \approx 0.25)\) of this compound. Here, we cover the influence of a frustrated bond in a centred tetragonal lattice of a DAF (e.g., \(Fe_xZn_{1-x}F_2\)) at \(H = 0\), for an extended range of frustration strengths in the entire interval of dilution \((0 \leq x \leq 1.0)\). \(M_S(x)\) is shown to be strongly dependent on the magnitude of frustration present in the whole range of \(x\). In particular it follows a percolation-like phenomenon characterized by an exponent \(\beta_p\). For frustrated samples a “long tail” effect is identified such that \(M_S\) is zero only at \(x_p\). On the other hand, \(Q(x)\) is almost independent on the frustration strength, except for \(x\) close to \(x_p\) where it suffers an abrupt increase in magnitude even for the case of a very weak frustration.

We use a microscopic Hamiltonian suitable to describe a randomly diluted Ising antiferromagnet, with short-range exchange interactions \(J_\ell\), given by:

\[
\mathcal{H} = \sum_{<i, \delta_\ell>} J_\ell \mathcal{E}_i \mathcal{E}_{i+\delta_\ell} S_i S_{i+\delta_\ell},
\]

where \(S_i = \pm 2\) (as in the \(Fe^{+2}\)), \(\mathcal{E}_i = 0, 1\) and \(\ell\) is summed up over the three sets of nearest neighbors in the centered tetragonal lattice shown in Fig. 1. We take \(N = 2 \cdot 30^3\) sites and use periodic boundary conditions. The local mean-field (LMF) method consists in solving iteratively the self-consistent equations involving the thermally averaged spins \(m_i\) obtained through a variational minimization of the MF-free energy\([7]\), which gives for \(\mathcal{E}_i = 1\)

\[
m_i = \langle S_i \rangle_T = 2 \tanh \left( \frac{1}{k_B T} \sum_{\delta_\ell} J_\ell \mathcal{E}_{i+\delta_\ell} m_{i+\delta_\ell} \right).
\]

The usual virtual crystal mean field (VCMF) result is obtained assuming \(m_i = m\) for every magnetic site. The LMF simulation starts by choosing a random initial configuration \(\{S_i, \mathcal{E}_i\}\) in the high-\(T\)(paramagnetic) phase. The system is cooled
in steps $\Delta T = 1K$ through (2) and heated back by the same amount up to a temperature of interest, following a convergence criterion:

$$\frac{\sum_i[(m_i)_n - (m_i)_{n-1}]^2}{\sum_i[(m_i)_n]^2} \leq 10^{-6},$$  \hfill (3)$$

where $n$ represents the $n$-th iteration. We measured the staggered magnetization

$$M_S(T, x) = (2/N) \sum_i \mathcal{E}_i m_i,$$  \hfill (4)$$

with $i$ summed over a given sublattice, and the spin-glass Edwards-Anderson (EA) order-parameter

$$Q(T, x) = (1/N) \sum_j \mathcal{E}_j < S_j^2 >_T,$$  \hfill (5)$$

with $j$ taken through the whole lattice. $M_S$ and $Q$ are averaged over 50 independent samples. In the simulations of the $Fe_xZn_{1-x}F_2$ compound we considered the exchange constants $J_\ell$, shown in Fig. 1, keeping unaltered the experimental ratios $j_1 \equiv J_1/J_2 = -0.013$ and $j_3 \equiv J_3/J_2 = +0.053$ measured[8] for $Fe_xZn_{1-x}F_2$, such as to fix the Néel temperature of the pure system, $T_N(x = 1) = 77.8$ K. This procedure appears to be quite realistic for application in $Fe_xZn_{1-x}F_2$, as early pulse experiments[9] in this system support no $x$-dependence of the total exchange interaction in the entire concentration range measured $(0.2 \leq x \leq 1.0)$.

The $x$-dependence of the normalized sublattice magnetization $M_S(x)/M_S(x = 1)$ is shown in Fig. 2, varying the frustrated interaction in the whole interval $0 \leq j_3 \leq 1$. Data were obtained at $T/T_N(x = 1) = 0.18$ and $H = 0$. The full line represents the expected result of the VCMF method for the specific case of $FeF_2$, where $j_3 = 0.053$. As well known, the VCMF method does not predict the existence of a percolation threshold. Moreover, in this method the presence of a weak frustrated interaction ($j_3 = 0.053$) plays no role in the linear dependence of $M_S$ with $x$. By way of contrast, in the LMF method we observe that even a very weak frustration ($j_3 < 0.053$) reduces dramatically the values of $M_S$ in the strong dilution regime ($x < 0.5$), as is
the case of $Fe_xZn_{1-x}F_2$ [3]. However, as can be seen from the $j_3 = 0.053$ curve, a weak frustration plays no effect on the AF order parameter $M_S$ of this compound for weak dilution ($0.6 < x < 1$). On the other hand, a strong frustration ($j_3 \approx 1$) may change the nature of the long range order even in the pure system, $x = 1$. $M_S(x)$ vanishes at the percolation concentration $x_p = 0.247 \pm 0.010$, determined by the projection to the $x$-axis of the best fit curve from the $j_3 = 0$ data. This is in very good agreement with the value $x_p = 0.243 \pm 0.010$, obtained[10] by series methods for non-frustrated b.c.c. lattices. In the LMF method $M_S$ becomes smaller than the VCMF values for $x$ below a frustration-dependent value $x(j_3)$. We perceive from the data of Fig. 2 that $x(j_3)$ increases for increasing $j_3$, but reaches the saturation value $x = 1.0$ for $j_3 < 1.0$. For non-zero frustration strengths the $M_S$ curves display tails toward the percolation threshold, after sudden initial drops starting at $x(j_3)$. Close to the percolation threshold, the antiferromagnetic order parameter have the asymptotic form $M_S \sim (x - x_p)^{\beta_p}$, where $\beta_p$ is the percolation exponent associated to the order parameter. The best fitting of this law, with $j_3 = 0$, gives the value $\beta_p = 0.63 \pm 0.11$. We notice the same behavior in the frustrated systems for which we find $\beta_p = 0.59 \pm 0.10$ for $j_3 = 0.053$ ($FeF_2$), and $\beta_p = 0.62 \pm 0.09$ for $j_3 = 0.265$. Series expansion[11] and renormalization group[12] methods yield values for $\beta_p$ between 0.4 to 0.5. One should stress, however, that $M_S$ vanishes only at $x = x_p$ for the whole interval $0 < j_3 < 1.0$.

The normalized Edwards-Anderson order parameter $Q(x)/Q(x = 1)$ is plotted versus $x$, for several values of $j_3$ in Fig. 3. The linear dependence of $Q(x)$ with $x$, found for $x > 0.4$, is not disturbed even for the strongest magnitude of frustration used in this work ($j_3 = 1.0$). However, close to the percolation threshold even an evanescent frustration, as is the case of $Fe_xZn_{1-x}F_2$, causes a pronounced increasing in $Q(x)$, as shown in the inset of Fig. 3. The values $M_S(x = 0.25) \approx 0$ and $Q(x = 0.25) \neq 0$
found in this LMF simulations for \( j_3 = 0.053 \) explains[3] the spin glass phase experimentally detected in \( Fe_{0.25}Zn_{0.75}F_2 \). For \( x < 0.24 \) however, both \( M_S \) and \( Q \) values are negligibly small (see Figs. 2 and 3, respectively) for the whole \( T \) interval measured in the present work (\( 2K < T < 150K \)). So, the LMF method does not support a true spin-glass phase (\( Q \neq 0, M_S \approx 0 \)) for very low concentrations of magnetic ions, in conformity with the short-range character of the exchange interactions.

In the LMF approach \( T_N(x) \) is determined by the inflection point of the \( M_S(x) \) versus \( T \) curve (see Ref. [3]). The \( x \) dependence of the normalized Néel temperature \( T_N(x)/T_N(x = 1) \), obtained by VCMF and LMF methods, is compared with experimental results for the \( Fe_xZn_{1-x}F_2 \) compound in the phase diagram of Fig.4. The LMF predictions for \( T_N(x) \) are in good agreement with experimental data in the compound \( Fe_xZn_{1-x}F_2 \) for the whole range of \( x \), in spite of the increasing error bars in the vicinity of the singular point \( x_p \). This large uncertainty is due to numerical difficulties to obtain \( M_S \) at very low temperatures (\( T/T_N(x = 1) < 0.17 \) in the strong-dilution regime \( (0.24 < x < 0.31) \). Notice that \( T_N(x) \) is virtually unchanged even by the presence of moderate strengths of frustration (see Fig. 2).

This is explained by the fact that in spite of a sudden reduction on the magnitude of the antiferromagnetic order parameter \( M_S(x) \) when a frustrated interaction \( (0 < j_3 < 1.0) \) is present, \( M_S(x) \neq 0 \) for all \( x > x_p \) (see Fig. 2). However, we cannot exclude the possibility that the inclusion of thermal fluctuations may destroy such tiny residual \( M_S(x) \), leading \( T_N(x) \) to vanish for \( x > x_p \), as experimentally observed in \( Fe_xZn_{1-x}F_2 \) [13].

Our reported results show that the presence of a small frustrated interaction plays no role on the magnetic properties of a DAF system for weak dilution. However in the moderately and strongly diluted regimes it causes major effects on a variety of magnetic properties, some of which have been identified in this work.
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Figure Captions

Fig. 1 b.c.c. magnetic structure, illustrating the first three nearest-neighbors exchange interactions between magnetic ions. 

Fig. 2 $x$-dependence of normalized sublattice magnetization $M_S(x)$, at $T/T_N(x = 1) = 0.18$ and $H = 0$, for various exchange ratios $j_3 = J_3/J_2$, measured by LMF simulations (symbols) and VCMF method (full line).

Fig. 3 $x$-dependence of normalized spin-glass Edwards-Anderson order parameter $Q(x)$, at $T/T_N(x = 1) = 0.18$ and $H = 0$, for various exchange ratios $j_3 = J_3/J_2$, measured by LMF simulations.

Fig. 4 $T_N(x)/T_N(1)$ vs. $x$ using VCMF (full line) and LMF simulations in the frustrated (●) and non-frustrated (□) cases, $H = 0$. Experimental data (△) of $Fe_xZn_{1-x}F_2$ are plotted for comparison (Ref. [13]). $x_p = 0.247 \pm 0.010$ indicates the percolation threshold.

\(^1\)Unfortunately we don’t have a ps file of this figure. However, it is also reproduced in Ref.[3].
\[ j_3 = 0.0 \]
\[ j_3 = 0.053 \ (FeF_2) \]
\[ j_3 = 0.265 \]
\[ j_3 = 0.795 \]
\[ j_3 = 1.0 \]

\[ H = 0 \]
$H = 0$
$\square j_3 = 0.0$
$\bullet j_3 = 0.053 \text{(FeF}_2\text{)}$
$\triangle$ experimental

$T_N(x)/T_N(1.0)$

$x$