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Spin arrangements in $CdCr_{2(1-x)}In_{2x}S_4$-type insulating re-entrant compounds

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Abstract. The spinel solid solutions $CdCr_{2(1-x)}In_{2x}S_4$ have been studied both by means of macroscopic magnetization measurements and by neutron diffraction in the dilution range $0 \leqslant x \leqslant 0.10$. The dramatic effect of frustration is evidenced, which leads to the disappearance of the ferromagnetic long-range order for 10% dilution. The $x = 0.05$ compound exhibits the characteristic features of re-entrance, but with a clear difference from metallic re-entrant systems. A qualitative 'local mean-field' model is proposed, which stresses the relevance of the range of the magnetic interaction.

1. Introduction

Spin-glass compounds have been the subject of numerous studies in the last 20 years, in connection with the great interest thus the physics of disorder has been giving rise to (for a review, see [1] and references therein). The first spin glasses to be widely studied were dilute solutions of magnetic transition-metal impurities in noble-metal hosts. In these materials, the exchange mechanism between the magnetic impurities is the oscillatory RKKY interaction; combined with site disorder of the impurities, it leads to a random distribution of coupling strengths and directions often associated with frustration. However, spin-glass phases were also observed in insulating systems, such as $Eu_{1-x}Sr_xS$, where random vacancies in the frustrated magnetic lattice can lead to a spin-glass state. For the metallic as well as the insulating systems, a paramagnetic-to-spin-glass phase transition occurs only for a sufficiently strong dilution and many questions still remain open concerning the weak-dilution domain. Although it was for a long time indicated by a question mark on the concentration–temperature phase diagram, this domain is now known to be associated with re-entrant compounds which, upon decreasing the temperature, first magnetically order and then develop a spin-glass-type frozen state. Until now, most of the attention devoted to this phenomenon has focused on the low-temperature phase the nature of which has given rise to controversy. Nevertheless, the relevance of the range of the magnetic interaction seems to have been rather well established now. The main features of re-entrance in metallic systems have been determined [2]; they consist of the coexistence, at low temperatures, of a long-range magnetic order of the longitudinal components of the spins with a spin-glass-like disorder among the transverse components. However, only very little is known about insulating systems, thus leaving room for contradictory points of view. In order to try to clarify this situation, we undertook a study of the insulating frustrated system $CdCr_{2(1-x)}In_{2x}S_4$, in the low-dilution limit [3]. Figure 1 shows the corresponding part of

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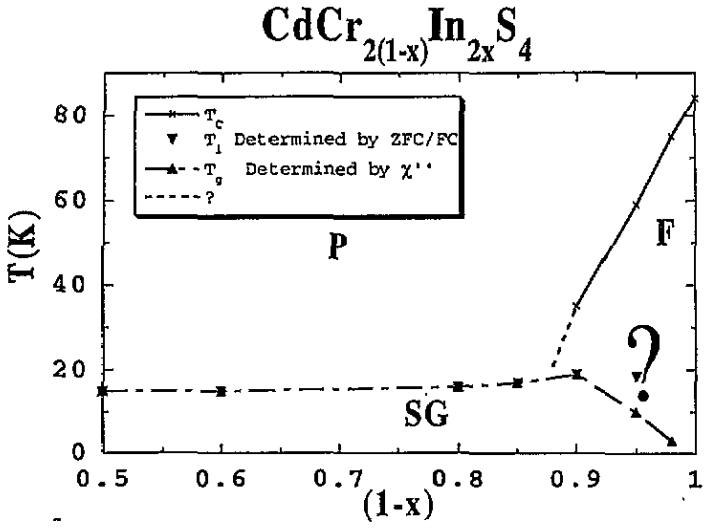

Figure 1. $CdCr_{2(1-x)}In_{2x}S_4$: low-dilution domain of the concentration—temperature phase diagram (from [4]).

its concentration–temperature phase diagram [4]. However, the authors particularly focused on the 'true spin-glass' phase [5], and this diagram is therefore not definitive.

When we started our investigation, we expected $CdCr_2S_4$ to be a 'good' three-dimensional Heisenberg ferromagnet, and this has proved to be true, as will be seen later. Its crystallographic and magnetic structures have been dealt with in a previous publication [6] and are briefly summarized here; $CdCr_2S_4$ is a normal spinel compound the Cr^{3+} magnetic ions of which occupy the octahedral B sites. Below $T_C \simeq 84$ K, long-range ferromagnetic order occurs, despite the existence of antiferromagnetic interactions between third-nearest neighbours, which remain unsatisfied, thus inducing frustration. Under these conditions, it is not surprising that a spin-glass state develops upon dilution. This process is furthermore favoured by the enhancement of the frustration due to the amplification of the antiferromagnetic couplings via the In^{3+} orbitals. This results in the absence of a percolating ferromagnetic cluster for dilutions greater than about 0.15.

We have performed both macroscopic magnetization measurements and neutron diffraction experiments on $CdCr_2S_4$ (to act as a reference) and the two dilute compounds $CdCr_{1.9}In_{0.1}S_4$ and $CdCr_{1.8}In_{0.2}S_4$, in order to characterize the different long-range ordered and re-entrant spin-glass magnetic phases.

2. Superconducting quantum interference device measurements

2.1. Experimental set-up

Low DC field ($H \leqslant 40$ Oe) magnetization measurements were carried out by means of a SQUID magnetometer, using an extraction method with a second-order gradiometer. The effects of residual magnetic fields were carefully taken into account and, in the cases of large signals (e.g. in the ferromagnetic phases) a calibrated flux attenuator was automatically switched on. The samples were polycrystalline powders ($m \leqslant 0.04$ g) enclosed in plastic

capsules. They were thermalized by a regulated helium gas flow and the temperature was measured with two thermometers; the temperature gradient along the 6 cm extraction path was always smaller than 0.2%.

For each compound, the magnetization measurements were performed by describing a hysteresis loop as a function of temperature. The sample was first cooled to the lowest probed temperature (about 8 K), without any applied magnetic field. The zero-field-cooled (ZFC) measurements were then performed by switching on the field at each selected temperature. When the highest temperature was reached (about 200 K), the sample was cooled again in the presence of the magnetic field. Field-cooled (FC) results were then obtained, by measuring upon heating the sample again.

In the low-field limit (which applies here), the response of the system can be taken to be a linear function of the excitation. Under these conditions, a measurement of the macroscopic magnetization directly gives access to the macroscopic susceptibility. The results obtained for the $x = 0.05$ and 0.1 compounds are presented in figure 2. As a comparison, the FC and ZFC susceptibility curves characterizing the $x = 0.15$ 'pure spin-glass' sample are also shown.

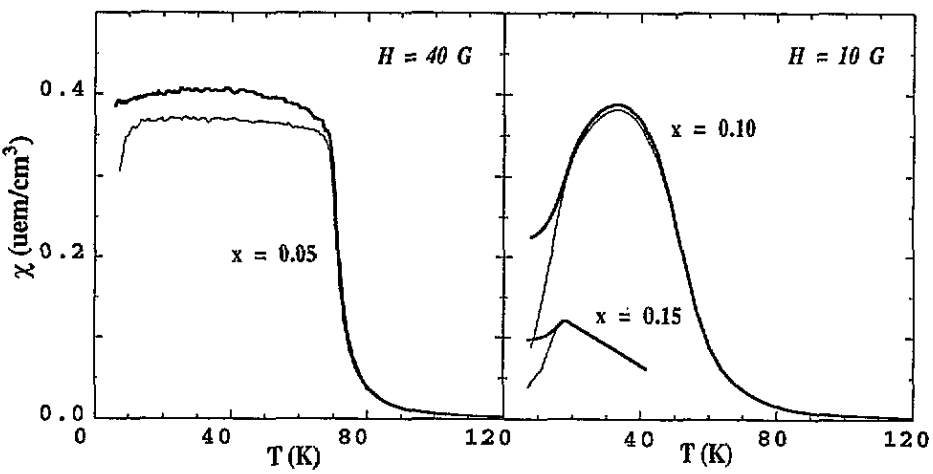

Figure 2. Temperature dependence of the FC (——) and ZFC (——) macroscopic susceptibilities, measured under a low magnetic field.

2.2. Experimental results

In the case of the pure $CdCr_2S_4$ compound, the temperature dependence of the macroscopic susceptibility is that of a 'normal ferromagnet'. When cooling from high temperatures, both FC and ZFC measurements lead to the same susceptibility value, which sharply increases close to 84 K. Then, below 84 K, the χ_{FC} and χ_{ZFC} curves separate. Both exhibit a plateau in the whole low-temperature phase (at least until 5 K), which, in the case of the FC susceptibility, corresponds to the constant value $1/N$, where N is the demagnetization factor. The samples were 2/1 ellipsoids and the corresponding value of $1/N$ is close to 0.4 emu cm^{-3}.

The introduction of 5% dilution only slightly modifies this ferromagnetic behaviour, leading to a decrease in the Curie temperature from 84 to 70 K. However, a new feature appears at low temperatures where, below about 10 K, the ZFC susceptibility starts to decrease.

The effect of dilution appears to be much greater for the $x = 0.1$ compound which exhibits a very peculiar behaviour. While on the one hand it does not look like a ferromagnet, the $\chi(T)$ curves being rather bell shaped, on the other hand, the susceptibility is much higher than that measured for the $x = 0.15$ spin-glass sample, reaching the $1/N$ limit value close to 32 K. The χ_{FC} and χ_{ZFC} curves clearly separate below about 17 K, after both have started to decrease.

In order to characterize further the different magnetic phases, we performed neutron diffraction experiments.

3. Neutron diffraction experiments

The samples used had a mass of 3 g each and were taken from the same batches as the SQUID samples. They were prepared from isotopic cadmium ^{114}Cd, and their neutron transmission, which of course depends on the degree of isotopic purity, was greater than 85% for the $x = 0$ and 0.05 compounds and close to 51% for the $CdCr_{1.8}In_{0.2}S_4$ sample. Experiments were performed on the D1B, E6 and G4.1 diffractometers, at the ILL, HMI and LLB reactors, respectively, always used in their standard set-up with typically $\lambda = 2.52$ Å and $10° \leqslant 2\theta \leqslant 90°$ for D1B. The collected data were analysed using the powerful FULLPROF Rietveld refinement program [7, 8].

3.1. Crystallographic structure

An important preliminary stage consisted of verifying that the crystallographic structure was not strongly modified by the dilution. This can be seen from figure 3, where the three high-temperature diffraction spectra are all very similar and are correctly fitted by the same structural model. The only change observed was an increase in the cell parameter with increasing dilution, which was expected from previous x-ray studies [9]. Nauciel-Bloch [10] reports $CdIn_2S_4$ to be partially inverse with 69% of In^{3+} ions on the A sites. He also gives an estimate of the degree y of inversion for the dilute compounds, the general formula of which can be written $(Cd_{1-y}In_y)\,[Cd_yIn_{2x-y}Cr_{2(1-x)}]S_4$, where ( ) and [ ] respectively correspond to A and B sites. For the refinements reported here, the degree of inversion was kept constant at the values reported in [10], i.e. $y = 0.05$ and 0.1 for $CdCr_{1.9}In_{0.1}S_4$ and $CdCr_{1.8}In_{0.2}S_4$, respectively.

3.2. Magnetic structure

For 5% dilution, magnetic Bragg peaks appear at around 70 K, which are characteristic of ferromagnetic order. The onset of this order is preceded by a strong increase in the diffuse scattering around the magnetic Bragg peak positions. As expected, this critical scattering diminishes when lowering the temperature below 70 K but then, below 30 K, the diffuse intensity increases again and passes well beyond the critical signal. This can be seen in figure 4, which shows the temperature dependence of the raw scattered intensity at about 2.5 'half-widths at half-maximum' (HWHMs) from the (111) Bragg peak position.

Performing a calibration of the recorded spectra with the nuclear scattering intensity, the Rietveld refinement program allows the determination of the apparent value M of the spontaneous magnetization from the integrated intensity of the magnetic Bragg peaks. The inset in figure 5 presents the thermal evolution of M, expressed in Bohr magnetons per Cr^{3+} ion, in the temperature range 1.4–70 K. The striking feature in this figure is the decrease in the spontaneous magnetization below about 20 K from a value of $2\mu_B$ to $1.7\mu_B$

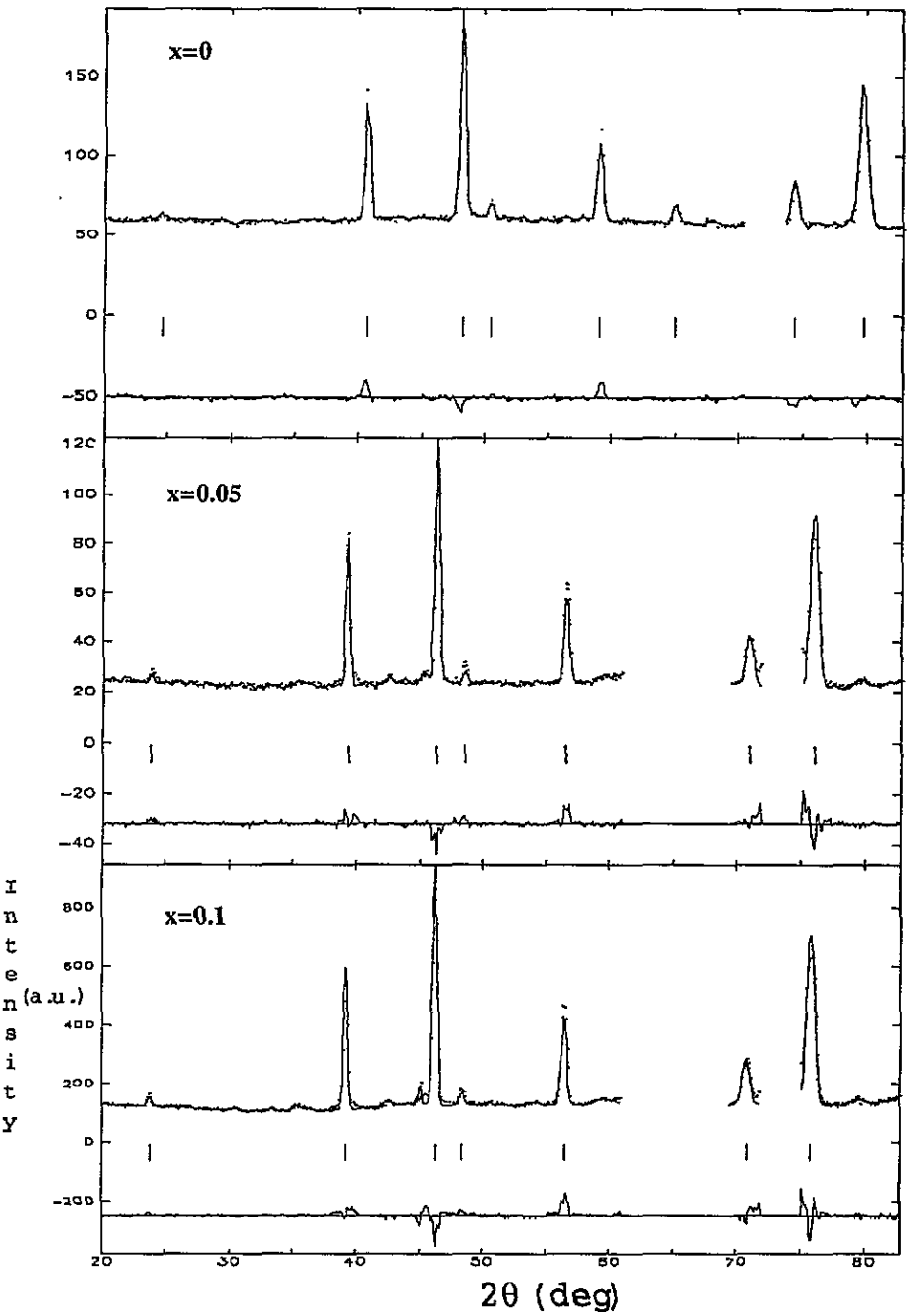

Figure 3. High-temperature diffraction spectra: comparison of the raw data with the calculated spectra obtained from Rietveld refinement (a.u., arbitrary units): $x = 0$, $\lambda = 2.52$ Å, $T = 180$ K; $x = 0.05$, $\lambda = 2.43$ Å, $T = 150$ K; $x = 0.10$, $\lambda = 2.43$ Å, $T = 110$ K.

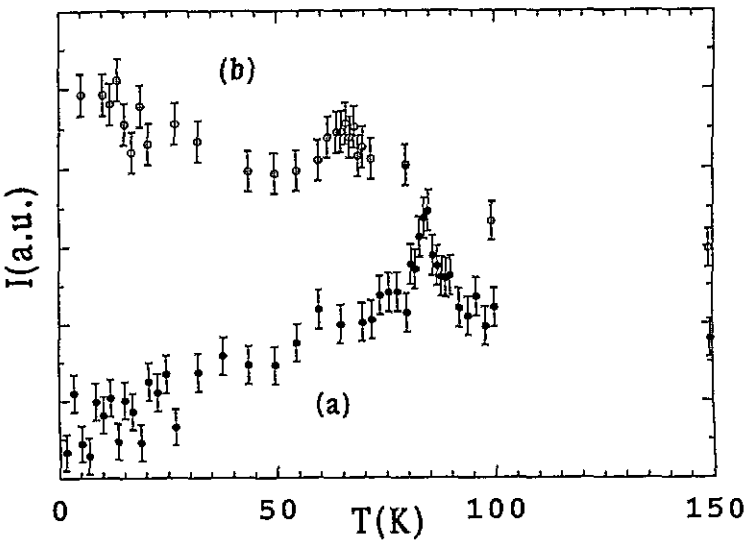

Figure 4. Temperature dependence of the raw scattered intensity, recorded at about 2.5 HWHMS from the (111) magnetic Bragg position (a.u., arbitrary units): (*a*) $x = 0$; (*b*) $x = 0.05$.

at 1.4 K. However, dilution appears in fact to reduce the apparent magnetic moment in the whole probed temperature range. Figure 5 shows the variation in the reduced magnetization $M(T)/M(0)$ as a function of the reduced temperature T/T_C, where $M(0)$ is the saturation value, equal to $3\mu_B$ for $x = 0$. In order to superimpose the two curves (at least in the 0.5–1 reduced temperature range), $M(0)$ must be set equal to $2.5\mu_B$ for $x = 0.05$. This represents a loss of 17% of the magnetization in the ferromagnetic phase. M starts to depart from the $x = 0$ curve at around 30 K, remaining rather constant at $2\mu_B$, and then diminishes below 20 K.

Before considering $CdCr_{1.8}In_{0.2}S_4$, some comments on the meaning of 'apparent magnetic moment' are essential. The partial coherent elastic magnetic cross section is customarily written in the form

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)^{magn}_{coh\ el} = N\frac{(2\pi)^3}{v}(\gamma r_0^2)F(Q)^2\exp(-2W)\langle S\rangle^2\left(\sum_\tau \delta(Q-\tau)\right)\delta(\omega)$$

where N is the number of magnetic scatterers involved in the percolating cluster, v the volume of the elementary cell, γ the gyromagnetic ratio of the neutron, r_0 the classical electron radius, $F(Q)$ the magnetic form factor, $\exp(-2W)$ the Debye–Waller factor and $\langle S\rangle$ the statistical average of the spin value. Thus, a change in the integrated intensity of the magnetic Bragg peaks can originate from either a variation in $\langle S\rangle$ (this is the case when varying the temperature in a normal ferromagnetic phase) or a modification of the number N of the spins which belong to the 'infinite' cluster. In this last case, the apparent spontaneous magnetization does not correspond to the order parameter of the ferromagnetic phase.

In the case of the more dilute $CdCr_{1.8}In_{0.2}S_4$ compound, the effect of dilution is very dramatic. No magnetic Bragg peak was observed in the whole probed temperature range, i.e. from 1.4 to 150 K, but diffuse scattering appears below 50 K. Its intensity, which is a maximum around 30 K, remains high at low temperatures, as can be seen in figure 4.

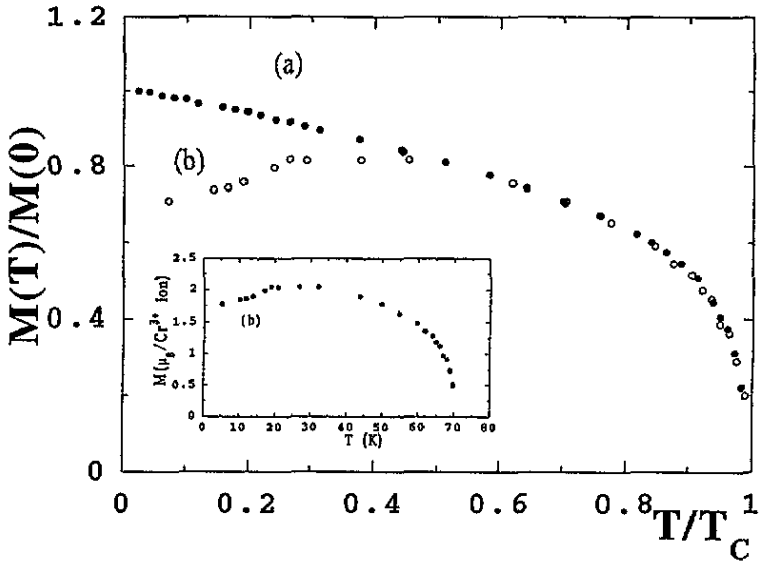

Figure 5. Reduced magnetization $M(T)/M(0)$ as a function of the reduced temperature. (*a*) For $x = 0$, $M(0)$ is equal to $3\mu_B$; this expected value was also deduced from the 1.5 K diffraction spectrum. (*b*) For $x = 0.05$, $M(0)$ was taken to be equal to $2.5\mu_B$.

3.3. Critical behaviour of the spontaneous magnetization

It is well known that the order parameter for the ferromagnetic phase is simply the spontaneous magnetization $\mu_B\langle S\rangle = M_S$. Below, but close to T_C, its variation as a function of the temperature is expected to follow the power law

$$M(T) = M_0[(T_C - T)/T_C]^{\beta} \tag{1}$$

for which the values of both the critical exponent β and the amplitude ratio $M_0/M(0)$ have been calculated in the framework of the renormalization group theory [11]. Experimentally, on the assumption that, close to T_C, the variation in the apparent magnetization M is only due to a change in the value of $\langle S\rangle$, the neutron diffraction experiments allowed us to determine the critical behaviour of the order parameter for both $x = 0$ and $x = 0.05$, and to compare the results with the theoretical predictions.

The investigation of the critical behaviour was done taking into account the (111) Bragg peak alone. Its nuclear component was determined from a high-temperature spectrum, by fitting to a linear background and a Gaussian peak. Close to T_C, one has also to deal with the strong critical diffuse scattering. However, since our main interest was in the diffracted intensity, this diffuse scattering was simply approximated to a Lorentzian function which, together with a linear background and a Gaussian function, was fitted to the raw data. In these adjustments, both peak centres were kept fixed, as well as the HWHM of the Gaussian. An example of one of these fits is shown in figure 6. We thus obtained the temperature dependence of the magnetization $M(T)$.

The second step consisted in adjusting the critical power law (1), which meant determining the value of the three parameters M_0, β and T_C. The Curie temperature was first estimated from the thermal variation in the intensity recorded at a constant scattering angle the distance of which from the first magnetic Bragg position was equal to 2.5 HWHMs; this led

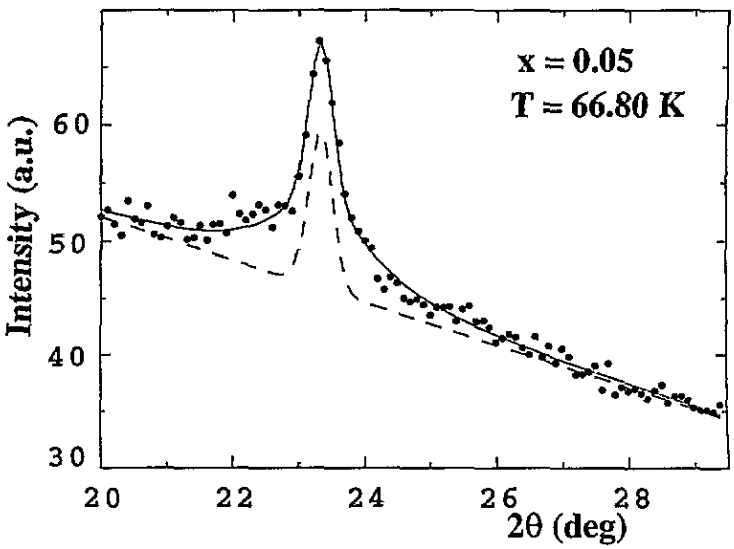

Figure 6. Scattered intensity around the (111) magnetic Bragg peak, at $T - T_C = 1.2$ K (a.u., arbitrary units): ——, line resulting from the adjustment of the three components linear, Lorentzian and Gaussian; – – –, the linear + Gaussian contribution.

to $T_C = 84.5 \pm 0.5$ K and $T_C = 68.0 \pm 0.5$ K for $x = 0$ and $x = 0.05$, respectively. We then adjusted the three parameters, restricting the acceptable values of the critical temperature to the pre-determined interval, and thus obtained the following results, illustrated in figure 7:

$$\begin{aligned} CdCr_2S_4 : & T_C = 84.1 \pm 0.2 \text{ K} \\ & \beta = 0.32 \pm 0.03 \\ & M_0 = 3.4 \pm 0.1 \mu_B/Cr^{3+} \text{ ion} \\ & M_0/M(0) = 1.13 \pm 0.08 \\ CdCr_{1.9}In_{0.1}S_4 : & T_C = 68.0 \pm 0.2 \text{ K} \\ & \beta = 0.30 \pm 0.03 \\ & M_0 = 2.8 \pm 0.1 \mu_B/Cr^{3+} \text{ ion} \\ & M_0/M(0) = 1.12 \pm 0.10. \end{aligned}$$

4. Discussion

Experimentally, the general features of re-entrance have been observed in various frustrated systems, such as Au_xFe_{1-x} [12], $Eu_{1-x}Sr_xS$ [13] and $Fe_{1-x}Cr_x$ [14]. These consist of first the onset of long-range order, followed by the occurrence of spin-glass-like properties. From a theoretical point of view, one can distinguish two main approaches to the problem, which are based on mean-field and random-field considerations, respectively.

The first was developed by Gabay and Toulouse [15] for the case of Heisenberg spins coupled through an infinite range interaction. They came to the conclusion that two

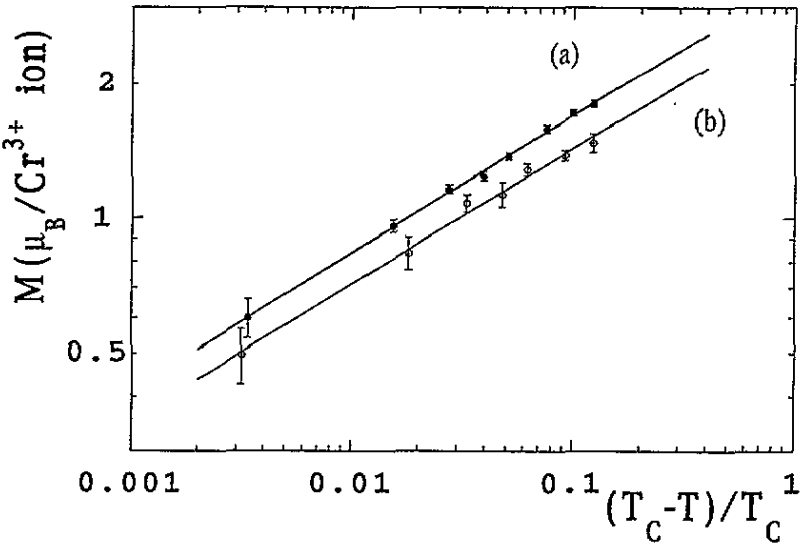

Figure 7. Critical law (1) adjusted to the $M(T)$ data; (*a*) $x = 0$; (*b*) $x = 0.05$.

characteristic temperatures existed below T_C. The highest characteristic temperature, T_1, was said to correspond to a spontaneous breaking of the replica symmetry in connection with the appearance of a mixed phase M_1 in which long-range order of the longitudinal components coexists with a spin-glass-type disorder in the transverse plane. The second temperature T_2 should be characterized by a transition towards a mixed phase M_2 with strong irreversibilities. It should be noted that, in both phases, the freezing of the transverse components is expected not to modify the longitudinal order.

These conclusions are in total contradiction with the predictions of the second model, based on an 'inhomogeneous' approach of the spin system. In this framework, the phase which develops just below T_C is described as consisting of both an ordered percolating cluster and finite ordered domains. At low temperatures, these domains are expected to freeze, thus inducing a random field which then breaks up the infinite cluster [16].

Until now, most of the experimental work on re-entrance has been performed on metallic systems. In this case, the mean-field approach of Gabay and Toulouse seems to be the most appropriate but nevertheless does not allow us to explain all the experimental observations such as the decrease in the longitudinal magnetization which occurs at low temperatures in AuFe. Studies of insulating systems have been performed on $Eu_{1-x}Sr_xS$, $Fe_{1-x}Mg_xCl_2$ [17] and very recently $Mg_{1+x}Fe_{2-2x}Ti_xO_4$ [18]. Shapiro *et al* [19] who carefully investigated the ferromagnetic order in a $Eu_{0.52}Sr_{0.48}S$ sample concluded that there was an absence of ferromagnetic long-range order in the re-entrant phase. This was presented as a corroboration of the random-field model predictions. However, it seems difficult to generalize such a result, which was obtained for a strongly dilute compound close to the percolation threshold (0.50 in this case). The main reason for this comes from the short range of the magnetic interaction which very probably enhances the effects of frustration and dilution, leading to a rapid change in the magnetic structure as a function of the amount of disorder.

Taking into account these considerations, we can now try to interpret our results. For both of the dilute compounds, the existence of a frozen magnetic phase at low temperatures is indicated by a decrease in the ZFC susceptibility and an enhanced remnant magnetization.

For the $x = 0.1$ sample, both of these features are distinct. The susceptibility curves show a maximum at the inverse demagnetization factor value, but no plateau. This compound is likely to be very close to the percolation threshold. In the case of $CdCr_{1.9}In_{0.1}S_4$, the spin-glass-type irreversibilities appear from a ferromagnetic state, when reducing the temperature. Owing to the finite instrumental resolution, it is difficult, from the diffraction data, to be sure of the presence of a truly percolating cluster. However, the combination of the Rietveld structure refinement results and the macroscopic measurements allow us to conclude that, at $T_C \simeq 69$ K, there is an onset of long-range ferromagnetic order. It should be noted that the departure from normal ferromagnetic behaviour does not occur at the same temperature, in both types of experiment. Although the magnetization measured by neutron scattering starts to decrease below about 20 K, the macroscopic susceptibility only departs from the ferromagnetic plateau below 10 K. This last feature has been observed in various other systems and has given rise to many controversies, since it could be explained in the framework of either a mean-field or a random-field theory. The reduction in the ZFC macroscopic susceptibility can indeed result from a blocking of the domain walls as well as from the disappearance of long-range order. Before trying to give an interpretation of our results, we should also mention the intermediate 'local-mean-field' model developed by Saslow and Parker [20]. In our opinion, this is the most suitable theory for insulating compounds. The starting point of the model consists in considering that, in the vicinity of a frustrated spin, the resulting local mean field from the neighbouring magnetic moments is weakened and cannot lead to a magnetic ordering process at T_C. Under these conditions, the frustrated spins remain paramagnetic and then, at low temperatures, freeze, thus exerting random forces on their ordered neighbours which may, as a result, separate from the rather tenuous percolating cluster.

The absence of diffuse scattering between T_C and about 30 K does not support the random-field model in which the percolating cluster and finite clusters should coexist in the ferromagnetic phase. However, on the other hand, the loss of 17% of the magnetization in the ferromagnetic phase cannot be explained within the framework of the Gabay–Toulouse theory. We therefore propose a qualitative inhomogeneous model for the spin system, which is also consistent with previous NMR results [21]. Investigating the low-temperature phase of very dilute $CdCr_{2(1-x)}In_{2x}S_4$ compounds, Méry *et al* concluded that, close to an In^{3+} impurity on a B site, the antiferromagnetic interaction is enhanced, leading to a local frozen canted disordered state around the lacuna. Owing to the short range of the interaction, one can think of this in terms of local mean fields which, close to an impurity, are weakened. Thus, in the spirit of the Saslow–Parker approach, it can be assumed that, even below T_C, the spins surrounding an In^{3+} ion on a B site cannot order and remain paramagnetic. This could explain the 17% loss of magnetization and the absence of diffuse scattering around the ferromagnetic Bragg positions, between 55 and 35 K. Then, upon cooling, these paramagnetic spins freeze in random positions, therefore exerting antagonist forces on their neighbouring ordered spins which can separate from the infinite cluster, alone or in finite clusters. This would lead to the observed increase in the diffuse scattering, and the reduction in the apparent magnetization, because of the smaller number of spins which belong to the infinite cluster. At low temperatures, the finite ordered clusters freeze in their turn and induce strong irreversibilities which appear as the decrease in the ZFC susceptibility. Unfortunately, our neutron diffraction experimental results do not allow us to determine whether or not the long-range order remains in the re-entrant phase. This comes from the finite spectrometer resolution which prevents us from distinguishing percolating from large (greater than 300 Å) ordered clusters.

In the framework of this approach, a small increase in the dilution is expected to

prevent the ferromagnetic order from percolating. $CdCr_{1.8}In_{0.2}S_4$ is likely to be very close to the percolation threshold since, with 10% of impurities, the probability for a spin to have a lacuna or a perturbed 'paramagnetic' spin in its close vicinity, which weakens its local mean field, is very high. Finite ferromagnetic clusters then develop and grow until they are nearly percolating close to 30 K. Then, the freezing of the paramagnetic spins prevents ferromagnetism from occurring and even reduces the ordered clusters, resulting in a decrease in both the FC and the ZFC susceptibilities. The finite clusters finally freeze at a low temperature (about 15 K), inducing an increase in the remnant magnetization.

In order to interpret the critical behaviour of the order parameter, it is tempting to refer to Brooks Harris' theoretical work. Dealing with the influence of disorder on the static critical properties, he was led to the conclusion—known as Brooks Harris' criterion—that, in the limit of weak disorder, dilution should be relevant only if the specific heat exponent α of the pure compound is positive [22]. $CdCr_2S_4$ is expected to be a 'good' 3D Heisenberg ferromagnet, which is in agreement with the value of the critical exponent β that we determined. This universality class is characterized by a negative value of α, equal to -0.126. Under these conditions, and assuming that Brooks Harris' criterion applies, the critical properties should not be modified by 5% dilution. This seems to be verified by our results. However do Brooks Harris' predictions apply in our case? In fact, this theoretical work was developed for a lattice of similar bonds, in which a small quantity of vacancies were introduced. It does not deal with frustration which, in our case, strongly enhances the disorder brought about by dilution. This problem appears to be more complex and certainly necessitates comparison with other theories, such as that developed by Heuer and Wagner [23], the aim of which was to extend Brooks Harris' results to the limit of strong disorder. This is beyond the scope of the present paper and will be the subject of a future publication.

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