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Induced ferromagnetic alignment in the one-dimensional Heisenberg linear chain antiferromagnet $(\text{CH}_3)_2\text{NH}_2\text{MnCl}_3 \cdot 2\text{H}_2\text{O}$ below the three-dimensional ordering temperature

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Abstract. We report on the magnetic effects induced by the substitution of Mn^{2+} ions in the quasi-one-dimensional Heisenberg-like antiferromagnet compound $(\text{CH}_3)_2\text{NH}_3\text{MnCl}_3 \cdot 2\text{H}_2\text{O}$ by non-magnetic Cd^{2+} and magnetic Cu^{2+} ions. In the undiluted compound three-dimensional ordering of the linear chains occurs at $T_N = 6.36$ K. In the diluted samples we observe the appearance of a ferromagnetic magnetization below T_N when the samples are cooled in small axial magnetic fields applied along the axis of antiferromagnetic alignment. At the lowest temperatures this magnetization is five times larger in the Cd diluted sample than in the Cu diluted one. Above T_N the susceptibility of both diluted samples shows an excess of magnetization with respect to that of the pure system. It is suggested that the uncompensated magnetic moments at the end of the 'magnetically broken' chains are responsible for this behaviour. It is also suggested that some of these 'free' paramagnetic ends can couple ferromagnetically at T_N , giving rise to the observed ferromagnetic component. The temperature dependence of the ferromagnetic magnetization of both samples is identical, indicating a common mechanism. The shape of this T -dependence suggests that the ferromagnetic correlations also have a low dimensional character.

1. Introduction

Recently it has been shown that some low anisotropy diluted anti-ferromagnets show a net spontaneous magnetization when cooled through their ordering temperature (T_N) in very small applied fields. So far this behaviour has been observed in diluted $\text{Mn}_x\text{Zn}_{1-x}\text{F}_2$, with $x = 0.25$ and 0.51 [1], in the series of three-dimensional (3D) antiferromagnets $\text{A}_2\text{Fe}_x\text{In}_{1-x}\text{Cl}_5 \cdot \text{H}_2\text{O}$ ($\text{A} = \text{K}, \text{Rb}$) for concentrations up to $x \sim 0.3$ [2, 3], and in diluted MnTiO_3 with Zn [4]. A similar behaviour was also observed in the diluted quasi-one-dimensional antiferromagnet $(\text{CH}_3\text{NH}_3)\text{Mn}_x\text{Cd}_{1-x}\text{Cl}_3 \cdot 2\text{H}_2\text{O}$ with $x \sim 0.01$ [5]. The spontaneous net magnetization appears along the easy axis of antiferromagnetic alignment when this axis is oriented parallel to the applied field (H_{axial}). In all these diluted systems, the moment at a given $T < T_N$, increases initially with H_{axial} but it saturates rapidly for H_{axial} of the order of 1 Oe. All these low anisotropy antiferromagnets have a ratio between the anisotropic and exchange fields (H_a/H_e) $\sim 10^{-3}$. As long as these diluted systems are cooled in the presence of the axial field, at a given temperature $T < T_N$, the magnetization can only be changed if the applied

field is greatly increased. In some cases fields of the order of 1 kOe are needed to reverse the direction of the magnetization. This remanent magnetization (M_r) increases with decreasing T . A remarkable characteristic of these compounds is that the temperature dependence of M_r below T_N seems to be x and compound independent [5–7], even for the system with marked one-dimensional character. The origin of this remanent moment remains unclear, but suggestions have been made that it may be due to an excess magnetization located at the walls of antiferromagnetic domains [6, 7]. The wall would be pinned by defects or by the non-magnetic sites.

The behaviour observed for the Heisenberg chain like system $(\text{CH}_3\text{NH}_3)\text{MnCl}_3 \cdot 2\text{H}_2\text{O}$ (MMC) diluted with Cd [5] deserves further comments. The magnetic susceptibility measured along the easy axis departs noticeably from that of the pure system even at temperatures well above the Néel temperature. This departure starts at temperatures below the temperature at which the broad maximum in the susceptibility– T curves occurs, T_M ($T_M \sim 23$ K for MMC). In one-dimensional systems T_M is governed by the antiferromagnetic exchange interaction within the Heisenberg chain. This temperature corresponds to the onset of the antiferromagnetic correlations along the chain. In the undiluted 1D system a three-dimensional ordering of these correlated chains occurs at a much lower temperature ($T_N = 4.12$ K for MMC). Since the introduction of impurities prevents the growth of the correlation length along the chain, the magnetic properties can be significantly changed. In solid solutions of $(\text{CH}_3\text{NH}_3)\text{MnCl}_3 \cdot 2\text{H}_2\text{O}$ with the Cd analogue (MMC/Cd) [5], an excess of susceptibility $\Delta\chi = \chi_{\text{diluted}} - \chi_{\text{pure}}$ with respect to the undiluted system is observed below T_M . The increase of $\Delta\chi$ with decreasing temperature follows a Curie law type behaviour as the 3D ordering temperature of the chains is approached. The extra susceptibility is due to an effective breaking of the magnetic correlation in the chains, which leaves extra ‘uncompensated’ moments at the end of half of the chains, e.g. those with an odd number of Mn ions. These moments could be responsible for the observed excess of magnetization. At the three-dimensional ordering temperature all these ‘free’ ends may correlate ferromagnetically giving rise to the observed spontaneous net moment. Since M_r appears at T_N , the sign of the inter-chain interaction may be of significance in this context.

In this paper we investigate another low dimensional system, $(\text{CH}_3)_2\text{NH}_2\text{MnCl}_3 \cdot 2\text{H}_2\text{O}$ (DMC) where Mn ions have been partially substituted by magnetic (copper) ions (hereafter denoted by DMC/Cu) and by non-magnetic (cadmium) ions (denoted DMC/Cd). As in the case of the previously investigated MMC, the manganese ions are in the same electronic state as those in the 3D systems previously studied, but here in a lower dimensionality structure. The novelty here is that we show that the effect of Mn substitution by Cd or Cu is very similar as concerns the behaviour of the remanent magnetization observed. This suggests that although the non-magnetic substitution is more effective in breaking the linear chains the magnetic copper ion may also generate an effective breaking of the magnetic one-dimensional correlations with similar consequences.

2. Experimental methods

The DMC is a Heisenberg linear chain antiferromagnet. A recent, structural refinement shows that it has an orthorhombic structure above 302 K and it is monoclinic below 302 K [8]. The chains are formed by groups of $[\text{Mn}-\text{Cl}_4-(\text{H}_2\text{O})_2]^{2-}$ octahedra linked by the chloride atoms along the c axis and are linked by hydrogen bonds in the b direction, forming planar sheets. In DMC and MMC, the di-methyl-amine and methyl-amine groups fill, respectively, the inter-planar space between the sheets. The chains correlate antiferromagnetically with an intra-chain interaction $J_{\text{intra}}/K = -2.7$ K below $T_M \sim 23$ K. At $T_N = 6.36$ K the chains

order three dimensionally with the magnetic moment perpendicular to the chains and parallel to the a^* crystallographic direction (a^* is perpendicular to the c - b plane) [9].

The three-dimensional (3D) ordering of this compound can be accounted for by an inter-chain exchange J' that is of the order of $10^{-2} J_{\text{intra}}$. The sign of this interaction is not reported in the literature. From its magnetic phase diagram [9], an estimate of the ratio of the anisotropy field to the exchange field, $\alpha = H_A/H_E = 3 \times 10^{-3}$, was found. This characterizes the compound as a low anisotropy Heisenberg one-dimensional (1D) system for temperatures above the 3D ordering temperature of the linear chains.

Magnetization and differential susceptibility were measured in samples of DMC/Cd, DMC/Cu and in pure DMC. Both magnetic measurements were made in a SQUID system manufactured by Quantum Design. The ac susceptibility measurements were made with an oscillating field of 1 Oe at frequency of 155 Hz. The 7 T superconducting magnet of the system provided the applied magnetic field. All samples were oriented with the easy axis parallel to the applied field. The field cycling curves (hysteresis curves) were taken with a vibrating sample magnetometer (VSM).

The diluted samples were prepared from a solution of manganese chloride and di-methylamine chloride in appropriate proportions. Controlled amounts of cadmium or copper chloride were added to the solution. The doping concentration, x , of Cd and Cu in the samples was determined by measuring the magnetization of the resulting diluted crystals in an applied field of 1 kOe. A sample of the pure system was measured also as a reference. In these measurements the temperature was varied from 50 to 290 K. Only the points above 150 K were used in the analysis. In this range of temperatures the magnetization curves could be well fitted to the Curie–Weiss law. From the Curie constants determined for each compound the concentration x of Cd and Cu was obtained. The shape and volume of the samples used were kept nearly the same. This was important to minimize differences in the magnetization due to filling factors. The values of x found in $(\text{CH}_3)_2\text{NH}_2\text{Mn}_{1-x}\text{A}_x\text{Cl}_3 \cdot 2\text{H}_2\text{O}$ are $x = 0.045$ for $\text{A} = \text{Cd}$ and $x = 0.06$ for $\text{A} = \text{Cu}$.

3. Results and discussion

The temperature dependence of the magnetization for DMC, DMC/Cd and DMC/Cu is shown in figure 1. For the three samples the magnetization measurements were made in an applied field of 100 Oe. The broad maximum associated with the correlations along the chain occurs at $T_M \sim 23$ K, the same value of T_M as obtained for MMC. In pure DMC, the magnetization curve $M(T)$ follows the expected behaviour of the parallel susceptibility ($M = \chi_{\parallel} H$) [9]. For the DMC/Cd and DMC/Cu samples there is a sharp increase in M below T_N . This is due to the remanent magnetization that appears at T_N in both diluted samples.

In the case of DMC/Cd a slight decrease in the ordering temperature is observed. If we take T_N as the point where M steeply increases we find $T_N(\text{Cd})/T_N(\text{pure}) = 0.99$. In contrast, the inclusion of Cu ions in DMC tends to increase the 3D ordering temperature: in this case we obtain $T_N(\text{Cu})/T_N(\text{pure}) = 1.01$. This temperature (T_N) is an effective measure of the coupling between the Heisenberg linear chains. Since the Cd inclusion decreases T_N and the Cu inclusion increases T_N we can conclude that for DMC/Cu the magnetic Cu ion may enhance the interactions between the chains. The effect of magnetic impurities in quasi-one-dimensional systems was explored in the antiferromagnet TMMC [10, 11]. In this compound the inclusion of either Cd or Cu impurities tends to decrease T_N , in contrast with the present case. The increase of T_N with the inclusion of Cu ions was also observed in MMC/Cu [5]. It is worthwhile to point out that within the TMMC chain the Mn–Cu impurity–host exchange (J_{IH}) is ferromagnetic whereas the host–host exchange (J_{HH}) is antiferromagnetic. No such

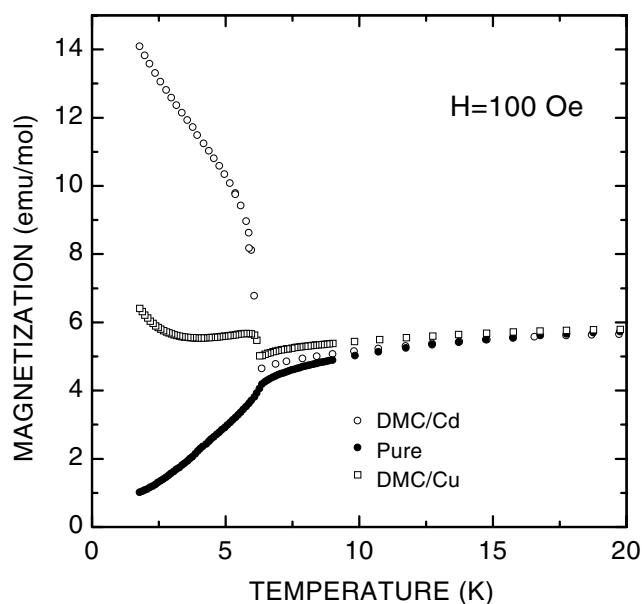


Figure 1. Measured molar magnetization at 100 Oe for the diluted samples DMC/Cd ($x = 0.045$) and DMC/Cu ($x = 0.06$). The remanent moment appears below T_N . The molar magnetization of the undiluted DMC compound is also shown for comparison.

study has been performed in either MMC/Cu or DMC/Cu. The three-dimensional ordering of impure linear chains was theoretically investigated in the case of non-magnetic and magnetic impurities [12]. For non-magnetic impurities a decrease in T_N is expected for both Ising and Heisenberg chains. For the classical Heisenberg model the effective change in T_N is linked not only to J_{IH} and J_{HH} but also to the inter-chain impurity–host (J'_{IH}) and host–host (J'_{HH}) exchanges. In that work a simple expression for this change with respect to the pure system is given:

$$\frac{T_N(x)}{T_N(0)} \approx 1 - \left(\frac{J_{HH}}{J_{IH}} - \frac{J'_{IH}}{J'_{HH}} \right) x \quad (J'_{IH} \neq 0).$$

We can readily see that with appropriate values and sign of these exchange parameters an increase in T_N is possible. Unfortunately only J_{HH} and the modulus of J'_{HH} are known for DMC and MMC.

In figure 2 the magnetization measured along the easy axis in an applied field of 1.0 Oe is shown for the DMC/Cd and DMC/Cu samples. At 2 K the total magnetization is $10.85 \text{ emu mol}^{-1}$ for the Cd diluted sample and $2.25 \text{ emu mol}^{-1}$ for the Cu diluted sample. Measurements made in fields of 0.5, 1.0, 2.0 and 3.0 Oe for the DMC/Cd sample show that the value of the magnetization at 2 K is nearly the same within 0.3%. This indicates that the remanent magnetization (M_r) appearing at T_N is already saturated at fields close to 1 Oe. Note in figure 1 and 2, that at the lowest temperatures the maximum value of the magnetization for DMC/Cd measured at 100 and 1.0 Oe, respectively, is of the same order of magnitude. The difference between the two values should be attributed to the small contribution of the magnetization due to the susceptibility, $\chi_{\parallel} H$, which at a field of 1 Oe is negligible compared with M_r and it is very small at a field of 100 Oe. This also occurs for the DMC/Cu sample.

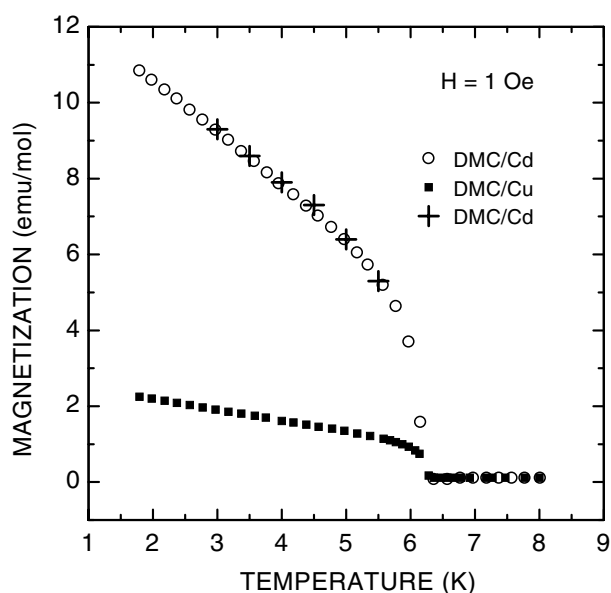


Figure 2. Molar magnetization for both diluted samples. The samples were cooled in the presence of a field of 1.0 Oe. The contribution to the magnetization of the parallel susceptibility $\chi_{\parallel}(T) \cdot H$ at 1.0 Oe is almost negligible. The data for the remanent moment at zero field as taken from the hysteresis curves (see text and figure 4) was also plotted (crosses) for DMC/Cd.

The measured AC susceptibility χ_{AC} for the DMC/Cd and DMC/Cu samples follows basically the behaviour of the static susceptibility $\chi_{\parallel} = M/H$ of the pure compounds (see figure 1). The static applied field in these measurements was of the order of 1 Oe. At T_N the measured susceptibility is close to $5 \times 10^{-2} \text{ emu mol}^{-1}$. Below T_N the appearance of the remanent magnetization in the diluted compounds is not revealed in the AC data because M_r is totally saturated at a static field $H \sim 1 \text{ Oe}$. The onset of the remanent magnetization is signalled only by the small peak structure seen in χ_{AC} .

The measured magnetization can be described by the relation $M(T, H) = M_r(T) + \chi_{\parallel} \cdot H$, where $M_r(T)$ is the remanent magnetization. The remanent magnetization in DMC/Cd measured at the lowest temperature ($\sim 11 \text{ emu mol}^{-1}$) corresponds to 0.08% of the magnetization (M_s) of the antiferromagnetic sub-lattice ($M_s \sim 14\,000 \text{ emu mol}^{-1}$). This is a much smaller proportion than that found in MMC/Cd where a 1% Cd dilution resulted in a remanent magnetization of the order of 80 emu mol^{-1} ($\sim 0.6\%$ of M_s) [5].

The behaviour of the system with Cu impurities shown in figure 2 for $H = 1.0 \text{ Oe}$ reveals that at concentrations close to that of the DMC/Cd system, the remanent moment M_r , although still large, saturates at much lower values (typically five times smaller). Another characteristic of the DMC/Cu compound is the presence of a non-negligible paramagnetic component that clearly manifests itself at the lowest temperatures in the magnetization curve taken at 100 Oe (see the data in figure 1 for DMC/Cu). Although in DMC/Cd a small paramagnetic component is also present, it is not so evident.

The appearance of a paramagnetic contribution in the magnetization curves for DMC/Cd and DMC/Cu shown in figure 1 is clearly seen below 20 K where the antiferromagnetic correlations within the chains are relevant. There is an excess magnetization in both diluted compounds as compared with the pure system. The paramagnetic contribution to

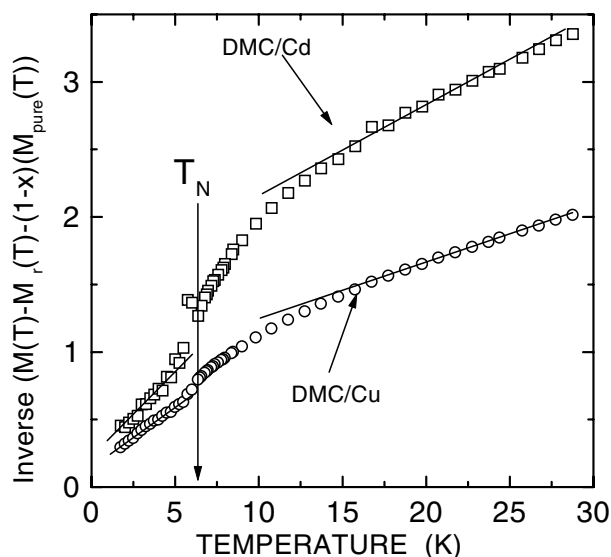


Figure 3. Reciprocal of the excess magnetization $[M(T) - M_r(T) - (1-x)\chi_{||}^*(T)H]$ at 100 Oe as a function of the temperature. Two Curie type linear behaviours are observed, one below T_N and the other above $T = 10$ K. From the slope of these linear regions we can conclude that some of the ‘free’ moments above T_N should participate in the observed ferromagnetic magnetization below T_N (see text).

the magnetization shown in figure 1 can be isolated from the data in both samples if the remanent magnetization that develops below T_N and the magnetization of the underlying antiferromagnetic structure is subtracted from the experimental magnetization. Since the remanent magnetization saturates at fields $H \geq 1$ Oe, the remanent magnetization $M_r(T)$ data below T_N shown in the curve of figure 2, can be subtracted directly from the magnetization data. To subtract the contribution of the antiferromagnetic structure the parallel susceptibility ($\chi_{||} = M/H$) of the pure DMC at 100 Oe (see figure 1) corrected for the fraction $(1-x)$ of Mn ions present in the diluted samples can be used. This last step supposes that only $(1-x)$ of the Mn ions contribute to the AF structure. The resulting curve is shown in figure 3 for DMC/Cu, and DMC/Cd. In this figure the inverse excess magnetization $[M(T) - M_r(T) - (1-x)\chi_{||}^*(T)H]^{-1}$ is plotted as a function of temperature. Two regimes of Curie type behaviour characterized by two distinct slopes in the plot are clearly observed. Below T_N in both diluted samples we observe a higher slope than that observed above 10 K. The crossover between these two regimes occurs between the 3D ordering temperature T_N and $T \sim 10$ K. In this region of temperature (~ 6 K $< T < \sim 10$ K), strong fluctuations due the proximity of T_N should be present. Since the slope is related to the inverse Curie parameter, an increase in slope for a given sample indicates a reduction in the number of ions that contribute to the ‘paramagnetic’ excess magnetization. Thus, some of the extra magnetic moments participating in the paramagnetic behaviour at higher temperatures are absent below T_N , where the remanent magnetization starts to develop. This result indicates that a fraction of these ‘paramagnetic’ ions correlate to give rise to the observed remanent moment. At this point it is relevant to emphasize that the correlation of these extra moments at T_N must lead to an effective ferromagnetic contribution, and for this a net ferromagnetic interaction is essential. This ferromagnetic interaction may be provided by the inter-chain exchange that can be ferromagnetic. Unfortunately the sign of this parameter is not reported in the literature

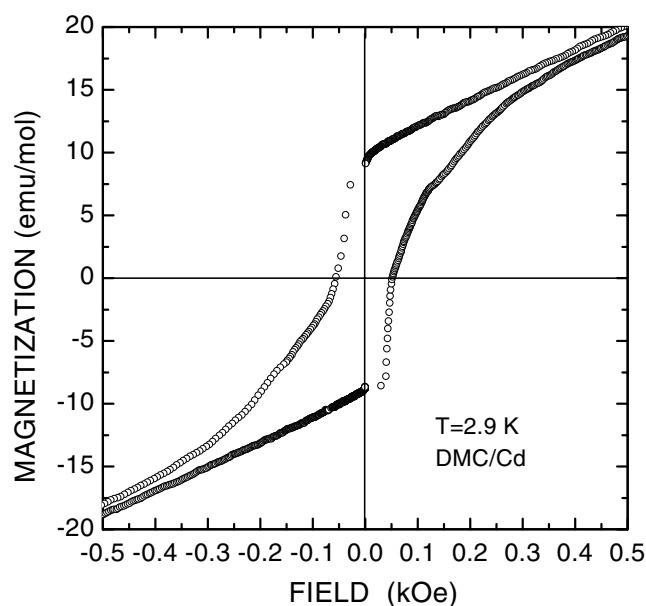


Figure 4. Hysteresis curve for DMC/Cd taken with the field parallel to the easy axis at $T = 2.9$ K. The sample was cooled to the measuring temperature in a field of 1.0 Oe. The values of M at $H = 0$ correspond to the remanent magnetization $M_r(2.9$ K). Similar points for $H = 0$ taken at several temperatures were plotted in figure 2 (crosses).

either for MMC or for DMC. Currently our group is investigating various low dimensional systems where the inter-chain exchange is well known in order to clarify this important point.

The coercive field below T_N can be estimated from the hysteresis data shown in figure 4. The magnetization of DMC/Cd upon a field cycling is shown there for $T = 2.9$ K. In these runs the sample was field cooled in a field ($H = 1.0$ Oe). The cycle was made between -10 and $+10$ kOe. The slope of the linear regions corresponds to the susceptibility of the antiferromagnetic underlying structure. Extrapolation of the linear region to $H = 0$ gives, as expected, the value for the saturated remanent magnetization at this temperature. The extrapolated values of the saturated remanent magnetization at different temperatures are plotted also in figure 2 with different symbols. The coercive field at $T = 2.9$ K is $H_c = 80$ Oe. Note that this coercive field is much higher than the field where saturation upon cooling through T_N is achieved (~ 1 Oe). The hysteresis curve of figure 4 also shows that a complete reversal of the moments below T_N occurs at fields exceeding 0.5 kOe at that temperature. This kind of behaviour suggests the gradual rotation of domains. A similar behaviour was also observed and discussed in detail in the three-dimensional antiferromagnets $\text{A}_2\text{Fe}_x\text{In}_{1-x}\text{Cl}_5 \cdot \text{H}_2\text{O}$ ($\text{A} = \text{K}, \text{Rb}$) [6]. The temperature dependence of the remanent magnetization for both samples is compared in figure 5. To represent these data we used the reduced temperature $t = T/T_N$ and the magnetization of both samples was normalized to unity at $t = 0.295$. The result clearly shows that the temperature behaviour is analogous for both samples suggesting that the underlying mechanism generating the ferromagnetic moment is the same. The temperature dependence found for these diluted DMC samples is very close to that already found in the other low anisotropy systems so far investigated [1–5].

One immediate guess about the origin of this remanent magnetization M_r can be traced to an imperfect compensation of the up and down sub-lattice magnetization. This point

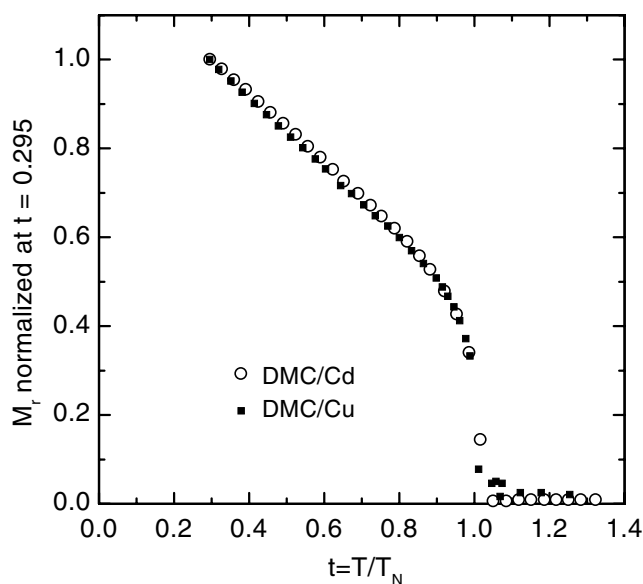


Figure 5. Dependence on the reduced temperature $t = T/T_N$ of the normalized magnetization $M(t)/M(t = 0.295)$ for the two diluted samples DMC/Cd and DMC/Cu. Note that both samples collapse into a single curve. A linear decrease of the magnetization with increasing temperature up to $t = 0.9$ is also observed.

has already been discussed in [1] and [2] where it was shown that if this were the case the temperature dependence of the remanent magnetization M_r should be identical to that of the antiferromagnetic sub-lattice. Unfortunately there are no direct measurements of the temperature dependence of the sub-lattice magnetization M_s either for MMC or DMC. An indirect comparison can be made however with the three-dimensional systems MnF_2 and $A_2FeCl_5 \cdot H_2O$ ($A = K, Rb$) for which the temperature dependence of the sub-lattice magnetization M_s is known [13, 14]. As already pointed out, the diluted $Mn_xZn_{1-x}F_2$, and $A_2Fe_xIn_{1-x}Cl_5 \cdot H_2O$ ($A = K, Rb$) show also a remanent magnetization whose temperature dependence is very close to that of MMC [5]. A direct comparison of the remanent M_r and the sub-lattice magnetization M_s carried out for these compounds shows that their dependence on temperature is very different (see for instance figures 6 and 7 of [3] where M_r and M_s are compared), ruling out the possibility that they could have the same temperature dependence. This in fact is the stronger argument against the picture in which uncompensated sub-lattices are at the origin of M_r .

It is remarkable, as already observed in the behaviour of the remanent magnetization of all other diluted low anisotropy compounds investigated [1–4], that the magnetization decreases linearly with increasing T in a wide interval of reduced temperatures. In the present systems the linear behaviour extends up to $t \sim 0.9$. A linear decrease of the magnetization with increasing temperature can be obtained within the mean field approximation for the ordered magnetic moments on the surface of an otherwise 3D system [15, 16]. This behaviour found in all the low anisotropy systems so far investigated may be an indication that the ferromagnetic correlations may have an effective low dimensional character, or even that this effect occurs at the surface of the samples or at the surface of antiferromagnetic domains. Magnetization measurements in ferromagnetic monolayers of Ni for example exhibit this kind of behaviour [17].

Although some of the results presented here give an idea of which moments are those that order ferromagnetically, the three-dimensional structure of this percolating network of correlated moments still remains unclear.

The fact that the pure sample does not show the ferromagnetic remanent moment links definitely the disorder in the magnetic lattice to the net moment. If plain surface effects of the sample alone were responsible the effect would be detected also in the pure system.

Acknowledgments

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