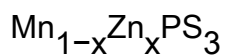


High-field magnetization in the diluted quasi-two-dimensional Heisenberg antiferromagnet



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High-field magnetization in the diluted quasi-two-dimensional Heisenberg antiferromagnet $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$

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Abstract

Magnetization of $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ in fields up to 35 T is presented. Small amounts of weakly bound moments have been observed for the more diluted compounds with a preferential orientation equal to that of the long-range antiferromagnetic order. The width of the spin-flop transition can be described by static domain wall broadening which is limited by the magnetic domain size. We estimate an average domain size of 200–250 nm.

1. Introduction

The magnetic behaviour of two-dimensional (2D) systems is of continuing interest as such systems are good testing grounds for fundamental magnetic theories. In particular, the study of the spin reorientation behaviour of antiferromagnetic compounds allows us to gain insight into the magnetic anisotropy and the different exchange interactions present within the material. Moreover, the diluted antiferromagnet is believed to be the experimental realization of the random field Ising model as was first realized by [1]. Random field effects induced by the application of an external field destroy the long-range order (LRO). This effect has been studied in detail with neutron scattering which reveals the disorder through the line shape of the magnetic scattering. Good examples include the 2D square Ising lattice $\text{Rb}_2\text{Co}_{1-x}\text{Mg}_x\text{F}_4$ [2] and the more magnetically isotropic $\text{Rb}_2\text{Mn}_{1-x}\text{Mg}_x\text{F}_4$ [3]. In the ordered state of the latter, the more diluted compounds show destruction of LRO before the compound enters the spin-flopped phase when an external field is applied.

In all these compounds, field cooling generates a metastable ordered state below T_N composed of microdomains. This state is destroyed upon entering the spin-flop (SF) phase where the moments are aligned perpendicular to the field. In this case the random fields do not couple to the order parameter and the LRO is recovered. Strong field-cooling effects are

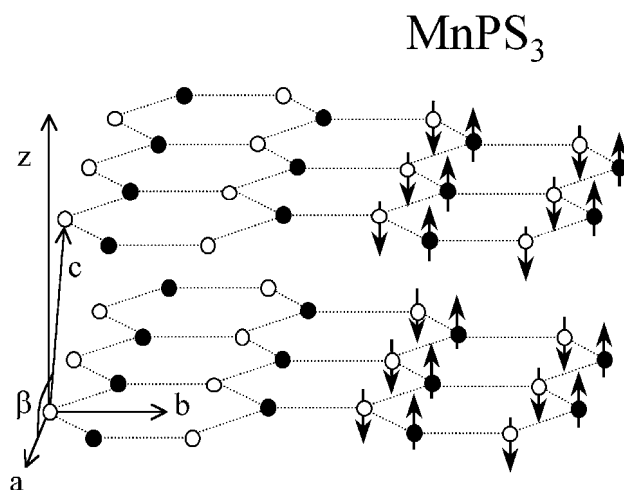


Figure 1. Honeycomb Mn(Zn) layers in $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$. The monoclinic crystal lattice (space group $C2/m$) has lattice parameters (for MnPS_3) $a = 6.077 \text{ \AA}$, $b = 10.524 \text{ \AA}$, $c = 6.796 \text{ \AA}$ and $\beta = 107.35^\circ$ [9]. The Mn nn distance equals 3.5 \AA . The antiferromagnetic structure of MnPS_3 below $T_N = 78 \text{ K}$ is illustrated, the black and white atoms corresponding to each magnetic sublattice [10]. The origin is arbitrary.

also seen in three-dimensional (3D) antiferromagnets such as $\text{Mn}_{1-x}\text{Zn}_x\text{F}_2$ [4]. If the sample is ordered in its microdomain state at low temperature and heated towards T_N , the Néel state is recovered for 2D systems well below the transition temperature, whereas for 3D systems this does not occur until the transition temperature is reached.

The SF transition, in which the moments flop from a parallel orientation with respect to the applied magnetic field to a perpendicular orientation, is broadened in 2D systems whereas it is sharp in 3D systems. Various mechanisms for the broadening in 2D systems have been suggested. Domain wall-like entities are thermally excited, as for example in the soliton model, and meander through the lattice [5]. Alternatively, for dimensions <2 , it is energetically favourable for domains larger than a critical size to break up due to random fields generated by imperfections in the lattice [6]. As this critical domain size increases with applied field, it has been suggested that the formation of additional domain walls occurs towards the SF transition [7]. On the other hand, SF transitions of 2D $\text{K}_2\text{Mn}_{1-x}\text{Mg}_x\text{F}_4$ in pulsed fields can be described by domain wall broadening due to the applied field only. In this case the magnetic dilution and consequent introduction of domain wall pinning centres does not introduce extra domain wall formation. It is believed this is due to the timescale of the experiment as there is insufficient time for relaxation.

In this paper we present high-field magnetization data on the $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ system and show how magnetic dilution with non-magnetic Zn atoms affects the SF and reorientation behaviour of the antiferromagnetic spins in applied magnetic inductions up to 35 T.

Within the $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ family of compounds, the Mn (and substituted Zn) atoms form a honeycomb lattice in the ab -plane and there is a relatively large distance between consecutive Mn(Zn) planes. The planes are separated by two layers of S atoms, which introduces a van der Waals gap. As a consequence, the exchange interactions within the planes are about 400 times stronger than those between the planes [8] and these compounds behave magnetically as quasi-2D systems. The crystallographic details and the magnetic structure are shown in figure 1.

The magnetic moment of Mn^{2+} ($S = \frac{5}{2}$) is solely of spin origin since the 3d electronic shell is half-filled. The absence of orbital magnetic moment causes the moments to behave isotropically with respect to their lattice. In the magnetically ordered state a small anisotropy is present ($S \parallel z$) due to the dipolar interaction between the spins. This classifies MnPS_3 ($T_N = 78$ K) as a 2D Heisenberg system with a small uniaxial anisotropy. Upon magnetic dilution by replacement of magnetic Mn^{2+} ions with non-magnetic Zn^{2+} ions, the breakdown of magnetic LRO is expected and the effects of the non-magnetic impurities in the antiferromagnetic lattice can be studied. Besides the introduction of random fields and the reduction in the number of magnetic atoms, the magnetic anisotropy is also reduced as it is of dipolar origin. All these effects jointly lead to the destruction of the LRO.

2. Experimental details

Samples of $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ were prepared with $x = 0, 0.07, 0.21$ and 0.45 using a vapour-transport method. Stoichiometric quantities of 99.99% pure elements were sealed in a quartz tube under $\approx 10^{-6}$ Torr of argon. The 18 cm tube was then placed in a two-zone furnace, with the temperature varying smoothly from 700 to 680 °C along the length of the tube. This produced a mass of transparent green crystals of hexagonal habit, typical dimensions being 5 mm \times 5 mm \times 0.1 mm. X-ray diffraction and microprobe analysis confirmed the structure and composition. Single crystals of $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ were cut into shape (diameter ≈ 3 mm) and stacked such that the z -axes were parallel but the ab -planes had arbitrary directions.

Magnetization measurements were taken at the High Field Laboratory in Amsterdam which permits semi-continuous magnetic inductions up to 35 T with a total duration of several hundreds of milliseconds [11] as well as at Monash University using a SQUID magnetometer in magnetic inductions of 0–7 T. The magnetic response of the Teflon sample holders was subtracted from all magnetization measurements. For each field pulse at Amsterdam, it was necessary to add a constant to the susceptibility to correct for uncertainties in the field compensation of the pick-up coil [11].

All measurements were taken at 4.2 K and with the applied magnetic field either parallel or perpendicular to the z -axis.

3. Dilution of the magnetic lattice

Within the antiferromagnetic state of MnPS_3 , the magnetic moments are aligned parallel to the z -axis, i.e. perpendicular to the honeycomb planes (see figure 1). This preferential orientation is primarily due to the magnetic dipolar interaction and the magnetic anisotropy energy K is therefore relatively small. A value of $K/k_B = 0.13$ K has been determined for MnPS_3 [12] and is considered a sum of a dipolar anisotropy ($K_{\text{dip}}/k_B = 0.16$ K) and a small single-ion anisotropy that favours an in-plane orientation of the Mn moments. This results in an anisotropy field of about 0.075 T.

In general, an ordered antiferromagnet prefers to align its moments perpendicular to an applied magnetic field ($\chi_{\perp} > \chi_{\parallel}$). If a field applied along the z -direction overcomes the magnetic anisotropy, the moments will flop into the plane at the SF field H_{sf} :

$$H_{\text{sf}} = \sqrt{\frac{2K}{\chi_{\perp} - \chi_{\parallel}}}. \quad (1)$$

Upon further increase of the applied field, the spins of both magnetic sublattices continue to rotate towards the applied field direction until saturation ($M = M_{\text{ord}}$) is obtained at

$$H_{\text{c},z} = 2H_{\text{e}} - H_{\text{a},z} \quad (2)$$

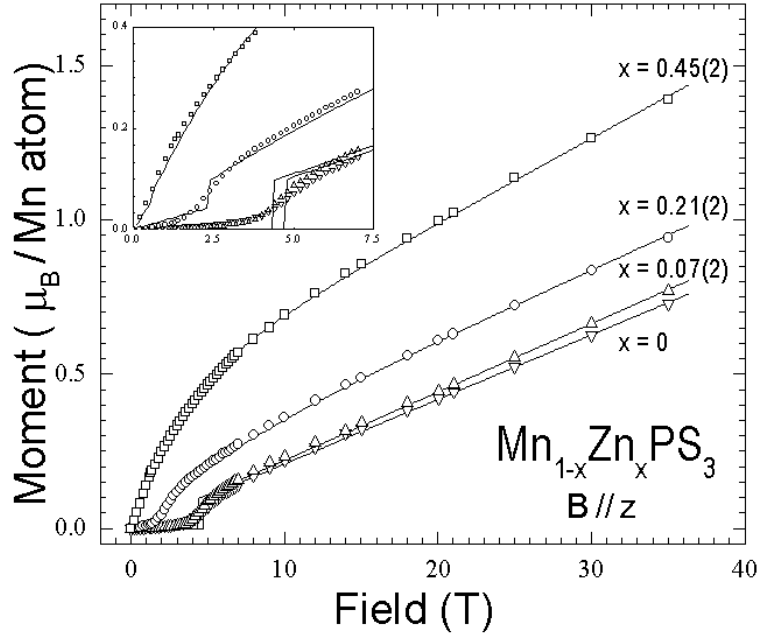


Figure 2. Magnetic response per Mn atom of the $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ series with $B \parallel z$. As the magnetic dilution increases, the SF field B_{sf} is strongly reduced due to the decrease in dipolar anisotropy (see the inset). The tilting of the moments towards the applied field direction after their SF reorientation is clearly visible in the linear behaviour above $B \approx 10$ T. Note that the reduction in exchange field is relatively small. At $x = 0.45$ (and $x = 0.21$) a weakly bound behaviour of some of the Mn moments is visible. The solid curves are fits as described in the text. The parameters deduced from these magnetization curves are listed in table 1.

Table 1. Selected magnetic properties of $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ for increasing values of magnetic dilution. The Néel temperature, T_N , indicates the onset of 3D magnetic order; the maximum observed in the susceptibility, T_{max} [13], is indicative of the onset of 2D magnetic order within the planes. M_{ord} is the ordered moment of the Mn atoms at 3.5 K measured by means of neutron diffraction [12]. The parameters are deduced from the experimental data in figure 2 in the following manner: $f_{\text{wb}} = M'(0)/M_{\text{ord}}$, $B_{\text{sf}} = (\frac{dM}{dB})_{\text{max}}$, $\chi_{\perp} = (\frac{dM}{dB})_{(10\text{T}-35\text{T})}/(1 - f_{\text{wb}})$ and $B_e = M_{\text{ord}}/2\chi_{\perp}$. Note that these values are calculated per Mn^{2+} atom within the $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ series.

x (—)	T_N (K)	T_{max} (K)	M_{ord} (μ_B)	B_{sf} (T)	χ_{\perp} ($\mu_B \text{ T}^{-1}$) atom Mn	f_{wb} (—)	B_e (T)
0(0)	78	120	4.5	4.8(1)	0.0209	—	108
0.07(2)	72	104	4.45	4.4(1)	0.0221	—	100
0.21(2)	54	T_N	4.32	2.4(1)	0.0231	0.03(1)	94
0.45(2)	20	T_N	2.86	0.6(1)	0.0303	0.09(1)	47

where $H_{a,z}$ is the anisotropy field in the z -direction and H_e the exchange field. Our specimens did not become saturated because H_e is considerably larger than the maximum pulsed field. Nevertheless, the increase of the moment along the applied field direction is linear and $H_{c,z}$ is deduced from its slope. The anisotropy field $H_{a,z} \ll H_e$ (at least a factor 10^3) and ignoring its contribution in equation (2) the exchange field $H_e = \frac{1}{2}H_{c,z}$.

High-field magnetization curves for $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ with $0 \leq x \leq 0.45$ as well as the low-field SQUID results are plotted in figure 2. We observe SF behaviour in all samples; the

value of $B_{\text{sf}} = \mu_0 H_{\text{sf}}$ decreases from 4.8 T at $x = 0$ –0.6 T at $x = 0.45$ (see the inset). μ_0 is the permeability of free space. For all samples, the relationship between the component of the Mn moment parallel to z and the external field was close to linear for $B > 10$ T. The width of the SF transition is broadened due to the 2D nature of the magnetic ordering [14]. For MnPS_3 and $\text{Mn}_{0.93}\text{Zn}_{0.07}\text{PS}_3$ the linear response extrapolates within error to (0, 0) indicating that all moments participate in the SF and reorientation behaviour. This is different for the $\text{Mn}_{0.79}\text{Zn}_{0.21}\text{PS}_3$ and $\text{Mn}_{0.55}\text{Zn}_{0.45}\text{PS}_3$ samples. The linear response extrapolates to a non-zero value of moment per Mn atom at $B = 0$. This suggests that some of the moments are already aligned parallel to the external field when the linear behaviour sets in and only the remaining moments contribute to the linear response observed above ≈ 10 T. The extrapolation of the linear response gives $M'(0) = f_{\text{wb}}gS$ where f_{wb} is the fraction of weakly bound moments. The parameters deduced from this simple observation as well as some known properties of $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ are listed in table 1. The exchange field of MnPS_3 is equal to that observed by [15] and is about twice the value as expected from the exchange constants ($J_1 - J_3$) determined by [8].

The solid curves in figure 2 assume a sharp SF transition and include a paramagnetic response for the weakly bound moments, with a certain effective Curie temperature, in the more diluted samples. In [16] the susceptibility of $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ has been successfully analysed with antiferromagnetic order and a Curie contribution from the uncompensated spins. Here we focus on the weakly bound moments.

From figure 2 it is clear that f_{wb} cannot be described by the Brillouin function, which predicts a significantly different low-field magnetic response compared to the measured values. This difference is particularly large for the $x = 0.21$ sample (inset: measured and predicted values). The increase in magnetic response of a paramagnetic system is largest at $B = 0$ whereas the derivative of the measured values is close to zero at $B = 0$. To gain insight into the nature of the weakly bound response we carried out a detailed analysis of the $x = 0.21$ specimen.

Figure 3 illustrates the low-field behaviour of $\text{Mn}_{0.79}\text{Zn}_{0.21}\text{PS}_3$ at 4.2 K. The magnetization was measured for $\mathbf{B} \parallel z$ and $\mathbf{B} \perp z$ and, since the anisotropy is small, the two curves merge in the region $B > B_{\text{sf}}$. $\mathbf{M} \perp z$ is analysed as the sum of a linear component representing the antiferromagnetic LRO (deduced from figure 2) and a non-linear component representing Mn moments that do not participate in the LRO. The presence of the linear component was inferred from the response for $B > 15$ T. In the region $B < B_{\text{sf}}$, the $\mathbf{M} \parallel z$ response is too small to include the same non-linear component, which suggests that all of the Mn moments are aligned in LRO. Thus, there are unlikely to be any truly free Mn magnetic moments in $\text{Mn}_{0.79}\text{Zn}_{0.21}\text{PS}_3$ at 4.2 K. We conclude that the weakly bound component is due to moments with a preferential orientation along the z -axis. A magnetic field perpendicular to z is able to tilt the moment and gives the weakly bound response (dotted curve in figure 3). On the other hand, a magnetic field parallel to the z -axis will not induce a moment reorientation but maintains the direction of the weakly bound moments during the SF transition. Consequently the weakly bound component is only present after the SF has occurred. For the $\text{Mn}_{0.55}\text{Zn}_{0.45}\text{PS}_3$ sample it is more difficult to determine the nature of the weakly bound moments as the difference between parallel and perpendicular susceptibilities is small. The solid curve in figure 2 is a best fit to the data and gives a paramagnetic Curie temperature $\theta = -7$ K. This suggests that in $\text{Mn}_{0.55}\text{Zn}_{0.45}\text{PS}_3$ also the weakly bound moments show a coupling to the LRO.

No differences between FC and ZFC magnetization curves have been observed [13] and the possibility of spin-glass behaviour as well as that of the presence of a ferromagnetic component can be excluded. This is as expected for a nearly isotropic 2D antiferromagnet. Yet, it also excludes possibility of the presence of random field effects in $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$. This is rather

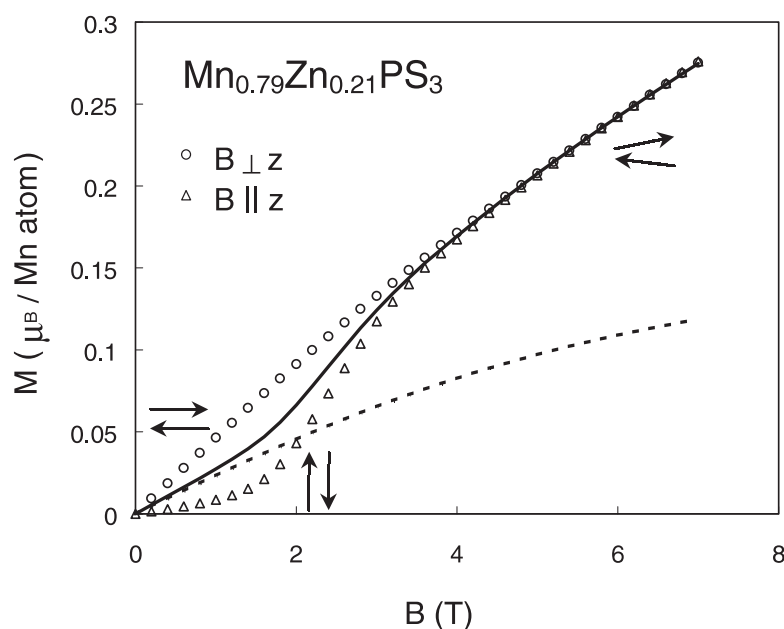


Figure 3. Magnetization of $Mn_{0.79}Zn_{0.21}PS_3$ for $B \parallel z$ and $B \perp z$. Note that the two curves merge for the spin-flopped state. For $B \perp z$ a non-linear response is observed which is supposed to be the sum of two components: a linear antiferromagnetic response whose signature is clearly visible in figure 2 beyond 15 T, and a weakly bound moment response drawn separately as a dotted curve. This latter curve represents the behaviour of Mn atoms that do not participate in the antiferromagnetic LRO. If these Mn atoms are magnetically free to orient themselves with respect to the field, their response would be the same for $B \perp z$ and $B \parallel z$. The solid curve represents the expected magnetic response including the spin-flopped component and the weakly bound component as determined from the $B \perp z$ magnetic response. The analysis is described in the text.

surprising and we attribute this to the fact that the exchange interaction is not dominated by nearest-neighbour (nn) atoms. The next nn and third nn contribute considerably and this reduces the influence of the random fields at the Zn sites. So although the honeycomb lattice is expected to be susceptible to dilution effects due to its small coordination number, this is less the case because of the strong beyond-nn exchange interactions in the plane.

4. Broadening of the spin-flop transition

The broadening of the SF transition in 2D systems with weak uniaxial anisotropy has been successfully described in the past by soliton (also called π -kink) excitations [5, 7]. A soliton corresponds to a domain wall which separates the two degenerate configurations of the AF lattice. The spins on each sublattice rotate simultaneously over an angle π within the domain wall whereby the direction of the two sublattices is interchanged. Just as for any other domain wall, the width of the soliton is a balance between exchange interaction and anisotropy. The solitons are excited thermally and require a minimum energy of creation. Any excess of thermal energy can be stored in the movement of the walls within the lattice. We used the formalism of [5] to analyse the SF transition in $MnPS_3$ within the framework of the soliton model. The results are shown in the left panel of figure 4.

For $MnPS_3$, the same response has been observed at 2, 6 and 10 K as can be seen in figure 4. This indicates that there are no thermal excitations of solitons or domain walls at

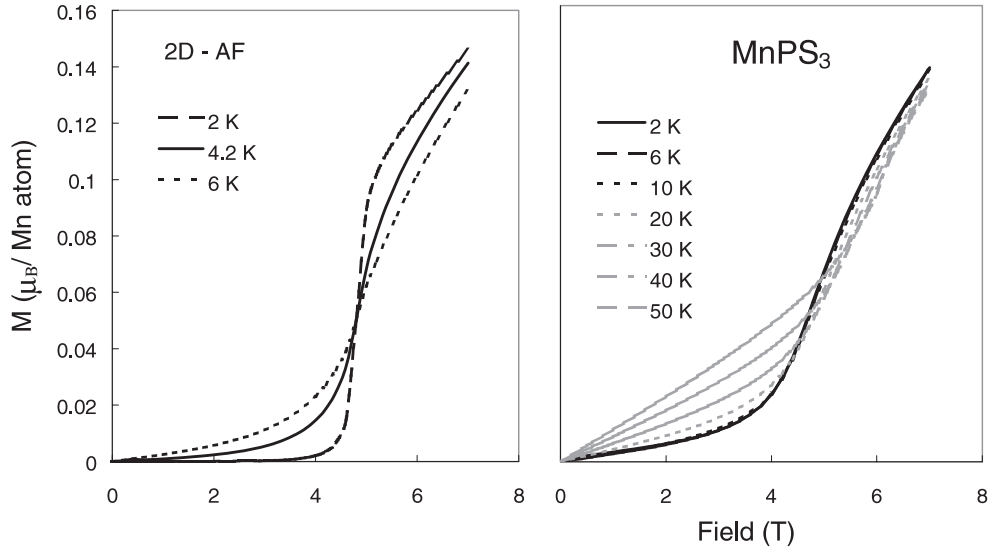


Figure 4. The SF transition predicted by the soliton model [5] (left) and experimental values at various temperatures in MnPS_3 (right). For the latter, the shape of the transition is temperature independent below ~ 10 K. B_{sf} increases slightly with temperature.

these temperatures since this would give a strong temperature dependence of the S-shape of the SF transition. It suggests that domain walls pinned at random fields produced by impurities or stacking faults broaden the SF transition as suggested by [6] and that no additional walls are formed during the SF transition. Below, we calculate the domain wall broadening due to the applied field and assume a constant number of domain walls.

The number of spins participating in a domain wall with width W is a balance between the exchange energy and the anisotropy energy. The first interaction drives towards infinite domain walls whereas the latter prefers a moment rotation of π between nn atoms. Assuming a one-dimensional (1D) problem and setting $\chi_{\parallel} = 0$, we estimate

$$E = J_{\text{ex}} S^2 \frac{\pi^2}{W} + \frac{1}{2} K W - \frac{1}{4} \chi_{\perp} H_{\parallel}^2 W \quad (3)$$

where the first term equals the exchange energy, the second term the anisotropy energy and the third term the Zeeman energy of the domain wall. H_{\parallel} is the applied induction along the z -axis of the MnPS_3 crystal and J_{ex} the nn exchange energy between spins S . The exchange interactions beyond nn are ignored here. A minimum in energy with respect to the domain wall width is found for

$$W^2 = \frac{4J_{\text{ex}} S^2 \pi^2}{2K - \chi_{\perp} H_{\parallel}^2} \quad (4)$$

This gives $W \approx 80$ atoms for MnPS_3 in zero field. The SF transition occurs as the Zeeman energy cancels the anisotropy energy and the domain wall width diverges. In a real system, W is limited by the average size of the magnetic domains D . When B is small, $W < D$ (figure 5(a)). W increases with B , so when $B = B_{\text{SF}}$ the domain wall is as large as the domain itself and $\chi = \frac{1}{2} \chi_{\perp}$ (figure 5(b)). Beyond the SF transition the domain wall, which is now the region in which the moments have a component aligned parallel to \mathbf{B} (perpendicular to the ab -plane), decreases in width, and the region in which the moments are perpendicular to \mathbf{B} grows at its expense (figure 5(c)). The use of symbols is indicated in figure 5 and the domain

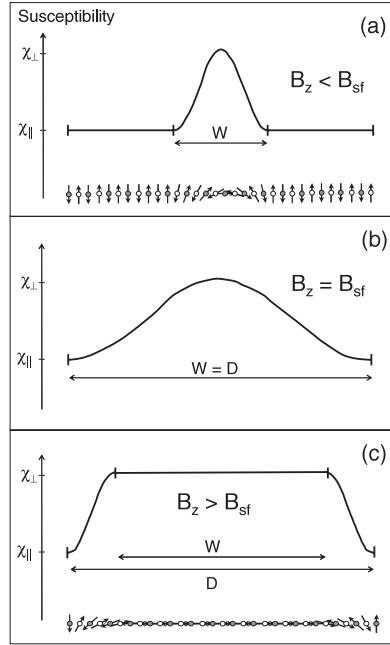


Figure 5. The magnetic response of the domain wall width W when a field is applied along the z -axis (a). At the SF transition the wall width is supposed to be limited by the magnetic domain size D in the material (b). At high field the magnetic moments are oriented perpendicular to the field except for moments in the domain wall (c). In reality the moments of the domain wall in (c) are also flopped into the plane where they are perpendicular to the moments in the region of width W .

wall width after the SF transition equals $D - W$. The energy of the system equals

$$E = J_{\text{ex}} S^2 \frac{\pi^2}{D - W} + \frac{1}{2} K W - \frac{1}{4} \chi_{\perp} H_{\parallel}^2 W. \quad (5)$$

We extended this model to a 2D system by simply assuming that moving domain walls are able to cross each other. In this case the 2D domain size is twice that of the 1D one for the same magnetic response. We are able to describe the SF transition in MnPS_3 with an average domain size of ~ 140 nm. Including the exchange interaction with the next nn and the third nn and treating it as an effective exchange interaction gives $D \sim 250$ nm. This corresponds to a distance between walls of about 900 atoms, a similar magnitude to that reported by [7]. Clearly, this calculation is a rough estimate and a strong simplification; nevertheless it indicates that the broadening of the SF transition can be attributed to impurities in the magnetic lattice only, combined with domain-size-limited domain wall growth.

A small change in gradient of about 5% has been observed in MnPS_3 at the SF transition when the field is applied perpendicular to z for various measurements at low temperature. Such a change can be expected if the preferential orientation of the moments is not perfectly along z or if part of the sample has a perpendicular orientation due to bending of the sample plates. Calculation of the dipolar energy for a collinear antiferromagnet on the MnPS_3 lattice gives an angle of 0.7° with the z -axis as the optimal orientation. The effect of this small angle on the perpendicular susceptibility is negligible; to account for the observed change, an angle of 13° with the z -axis is required. Alternatively, 5% of the sample should be perpendicular to the field to account for the observed effect. Both are larger than can be expected from the mounting of our samples.

On the other hand, a SF transition of the domain walls which have moments parallel to the applied field could account for the change in gradient. On average, half of the domain walls have their moments parallel to the applied field if we assume that there is no anisotropy within the ab -plane. These walls could flop perpendicular to the field at the SF transition, thereby becoming identical in orientation to the other half of the domain walls. Assuming a natural domain wall width of 80 atoms (equation (4)) we calculate a domain size of 230 nm. This is in rough agreement with our previous estimate and is a lower limit if we include the experimental uncertainties mentioned before.

As a consequence, the model as drawn in figure 5(c) is incorrect. The moments of the domain walls in the SF phase are flopped into the plane where the moment direction is perpendicular to that of the moments within the domain. The effect of this on the estimated domain size is small and included in the values mentioned earlier.

SQUID measurements are taken with a slow timescale which allows relaxation to occur. The magnon gap equals 0.5 meV [8] and relaxation is expected to take place in the 10 K measurement but less so in the 2 K measurement. As the SF response in MnPS_3 is equal for the measurements between 2 and 10 K, the formation of extra domain walls during the SF transition is not likely.

5. Conclusions

As the magnetic dilution is increased in the $\text{Mn}_{1-x}\text{Zn}_x\text{PS}_3$ series, we observe a strong reduction in SF field and a reduction in exchange field. Even for the $x = 0.45$ sample, which is beyond the percolation limit of the honeycomb lattice, all spins seem to participate in the LRO, although a proportion of the moments (9%) are weakly bound. For the $x = 0.21$ sample we clearly observe that 3% of the moments are weakly bound to the LRO and show a preferential orientation along z . This illustrates that the beyond- nn exchange interactions in the plane play a significant role which is confirmed by the absence of random field effects in these compounds.

The broadening of the SF transition in MnPS_3 has been described by domain-size-limited domain wall growth. We estimate an average domain size of 200–250 nm. There are no indications that extra domain walls are generated during the SF transition.

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