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Evidence for a new magnetic phase in MnP at very low fields

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Abstract. A new magnetic phase was observed in MnP at temperatures below its Curie point (292 K). A sharp increase in the in-phase and out-of-phase components of the AC susceptibility is observed at $T = 282$ K when fields below 5 Oe are applied along the intermediate anisotropy (***b***) axis. The new magnetic phase is observed down to 60 K where it gradually merges with the previously known ferromagnetic phase. Based on the AC studies performed we propose that this phase corresponds to a spin reorientation transition.

1. Introduction

The magnetic phase diagram of manganese phosphide (MnP) exhibits several ordered phases, including different kinds of modulated phase. It is the only magnetic system in which a pure Lifshitz type critical behaviour [1] has been found and critical exponents measured [2–4]; evidence of the existence of a region of commensurate–incommensurate transitions in the phase diagram was also given [5]. Various types of critical point have also been found including a line of multicritical Lifshitz points [6]. The phase transitions and the observed critical points can be simply explained in terms of the competition between ferromagnetic and antiferromagnetic interactions in the system [7]. The phase diagram of MnP was extensively explored for various directions of the applied field. The 3D phase diagram of MnP is schematized in figure 1. We refer to previous papers for a detailed discussion of the richness of phases and the different types of critical behaviour encountered in this material. At $H = 0$ the ferromagnetic phase is observed between 47 K and the Curie temperature $T_C = 292$ K. Below 47 K a modulated phase where the magnetic moments have a 'screw' spin configuration is observed at $H = 0$. In this paper we report two new features that were found in the region close to the Curie temperature (T_C) of this system: (1) the existence of a new phase transition 10 K below T_C and (2) the observation of a maximum in the AC susceptibility in the paramagnetic region.

2. Experimental results

The AC magnetic susceptibility of two single crystals on MnP oriented with the applied field along the intermediate b axis was measured with a Quantum Design SQUID magnetometer. One of the crystals consisted of a rectangular slab ($m = 54.6$ mg) with ∼0.7 mm thickness and sides of ∼4 by ∼5 mm^2. The crystallographic ***b*** axis was along the side of 4 mm. The other sample consisted of a spherical crystal ($m = 444$ mg) with a diameter of 5.3 mm. These crystals came from different sources. The AC measurements were made at the frequencies of 1.0, 10.0, 100.0 Hz and 1 kHz. The modulation field (h) was set at 2 Oe and it was always

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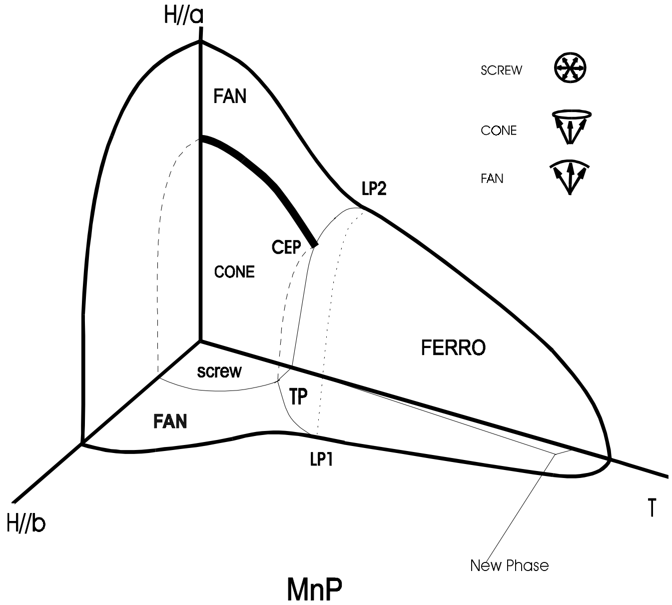


Figure 1. Sketch of the magnetic phase diagram of MnP in the $H \parallel a$, $H \parallel b$–T space. The dotted line at $T \sim 120$ K links the Lifshitz points (LPs) that occur when H lies along the $\boldsymbol{a}$ and $\boldsymbol{b}$ directions. The dashed line ~45 K connects the critical end-point (CEP) observed when $H \parallel a$ with the triple point (TP) in the phase diagram with $H \parallel b$. Transition between the ordered phase and the paramagnetic phase are second order. The transitions between the ordered phases are first order except for the cone–fan boundary for $H \parallel a$ that is second order and ends at a CEP. Sketches of the magnetic moment configuration of the modulated phases are given; they should be viewed as the projection of the moments in successive $\boldsymbol{bc}$ planes. The region between 47 and 282 K close to $H_b = 0$ indicated in this figure corresponds to the new phase observed for the first time in this paper.

applied in the same direction as the applied static field (H). All the data shown in the figures are for the slab sample. Both samples are compared in the inset of figure 3(a). Figure 2 shows the measured curves for the slab at several values of the static field ranging from $H \sim 0$ to $H = 10\,000$ Oe and applied along the intermediate anisotropy $\boldsymbol{b}$ axis. One prominent feature of these curves is the huge increase in the susceptibility at $T \sim 282$ K for the curve corresponding to $H \sim 0$. The results for the spherical sample are similar. Analogous measurements made with the fields h and H applied along the hard $\boldsymbol{a}$ axis do not show this kind of behaviour at $H \sim 0$. In figure 3(a) the in-phase (χ') and out-of-phase (χ'') component at $H \sim 0$ are shown for temperatures between 25 and 300 K. This figure gives an overall view of the whole region in temperature where the ferromagnetic phase of MnP is observed, namely between 47 and 292 K. The measurements shown in figure 3(a) were taken with an applied field very close to $H = 0$: we estimate that this field is under 0.3 Oe. Note that the out-of-phase component, related to the losses in the system, have a non-negligible value down to temperatures close to the ferromagnetic–screw transition at 47 K. In the inset of this figure the results for χ' of the slab and the sphere are compared. The difference in the values of χ' below $T_C = 292$ K is due to the distinct demagnetization factor of the slab and the sphere. The χ' and χ'' susceptibilities were also measured in static low fields up to 5 Oe. These data are shown in figure 3(b). Note the gradual decrease in the jump at $T \sim 282$ K with increasing applied field. Figure 4 exhibits the frequency dependence of χ' and χ'' in the region near the new transition. Frequencies were set at f = 1, 10, 100 and 1000 Hz and the applied static field H was close to 1 Oe.

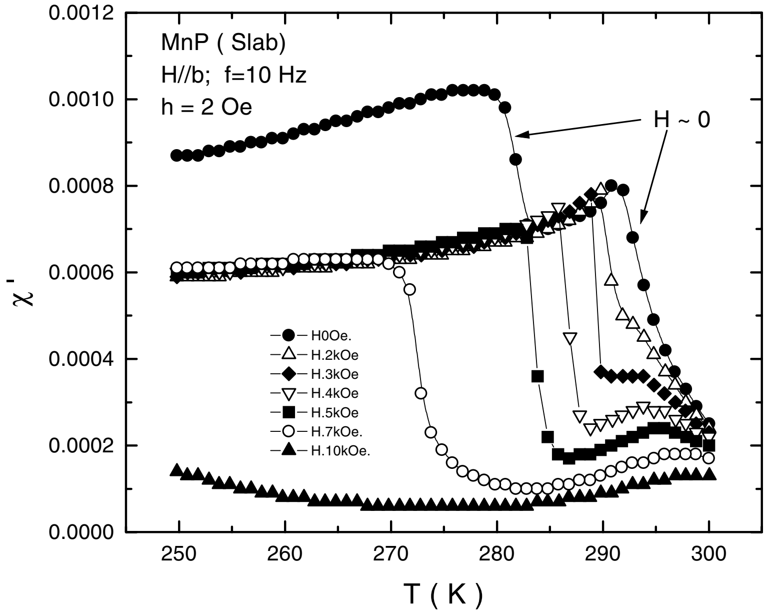


Figure 2. Susceptibility χ'–T curves for different fields up to 10 kOe applied along the b axis. Note at $H \sim 0$ the sharp increase in the signal at $T \sim 282$ K. This point marks the onset of the new observed transition.

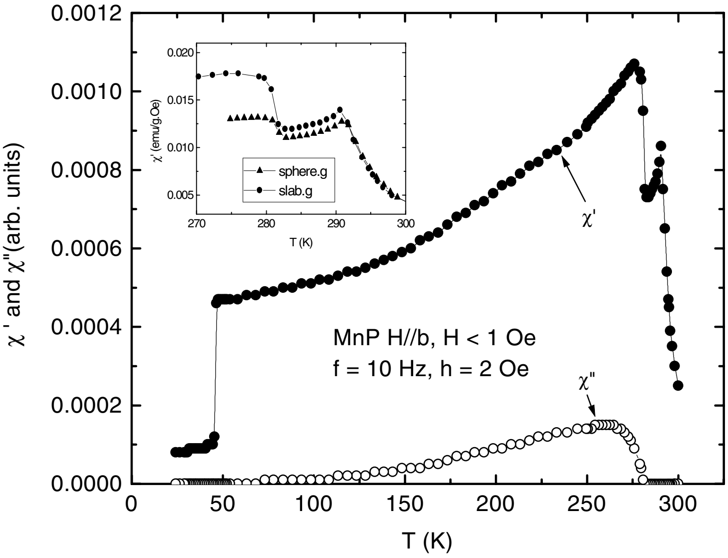
3. Discussion

Figure 1 is a sketch of the observed phase diagram of MnP in the T–field space, based on the data reported for this system [6]. The relevant portion of this diagram for this paper is the one that encompass the ferromagnetic phase that occurs above the screw–ferromagnetic transition at 47 K and at $H = 0$. At zero field the system orders ferromagnetically at 292 K with the magnetic moments aligned along the $\boldsymbol{c}$ axis of the crystal, the crystallographic $\boldsymbol{b}$ and $\boldsymbol{a}$ axes being respectively along the intermediate and high anisotropy directions. For higher fields $T_C(H)$ decreases with H as is sketched in the phase diagram of figure 1. The phase boundary $T_C(H)$ decreases faster when H is applied along the $\boldsymbol{b}$ axis since this corresponds to the intermediate anisotropy axis. This is the direction where we applied the field in this work.

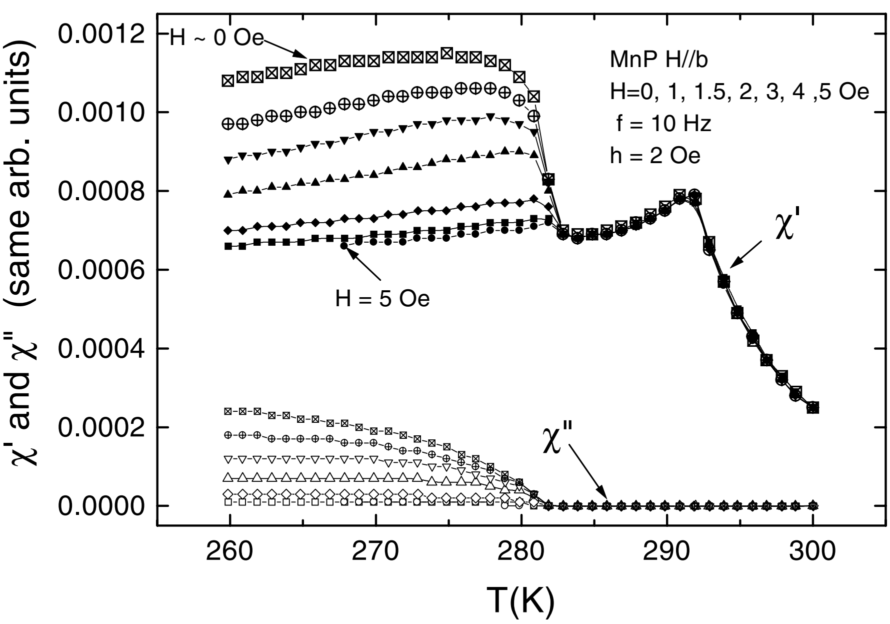
At this point let us summarize the main observations that can be obtained from the experimental data exposed in the previous section:

(1) From figure 2, which extends in temperature up to 300 K, we can observe that above the paramagnetic–ferromagnetic transition temperature (T_C) a maximum in the susceptibility is observed at a temperature $T_m > T_C$. This maximum is more evident in the curves obtained at higher fields where the decrease in the transition temperature $T_C(H)$ due to the applied field is more pronounced (see figure 1 for the shape of this phase diagram in the T–field space). The temperature where this maximum occurs increases with increasing field while the transition to the ferromagnetic phase decreases. In fact this maximum appears also when the external field is applied along the easy (c) axis (not shown).

(2) From figure 2 we also observe that at $H = 0$ a sharp transition occurs around $T = 282$ K, roughly ten degrees below the Curie temperature. This transition is completely suppressed by the field applied along the b axis, as can be seen in the curves measured at higher fields.



(a)



(b)

Figure 3. (a) Susceptibility χ against T obtained with a modulation field of 2 Oe and an excitation frequency of 10 Hz. Both the in-phase χ' and out-of-phase χ'' component of χ are plotted. Note the sudden increase in χ''. In the inset χ' is compared for the slab and the spherical samples (no correction for demagnetization effect was applied). (b) The effect of static applied fields up to 5 Oe is shown between 260 and 300 K. Note that a field of 5 Oe almost suppresses the anomaly at 282 K.

Only the ferromagnetic transition at $T_C(H)$ is present in these cases. In figures 3(a) and 3(b) we can observe that this new transition is accompanied by a sudden increase in the out-of-phase component (χ'') of the susceptibility. This non-negligible value of χ'' extends

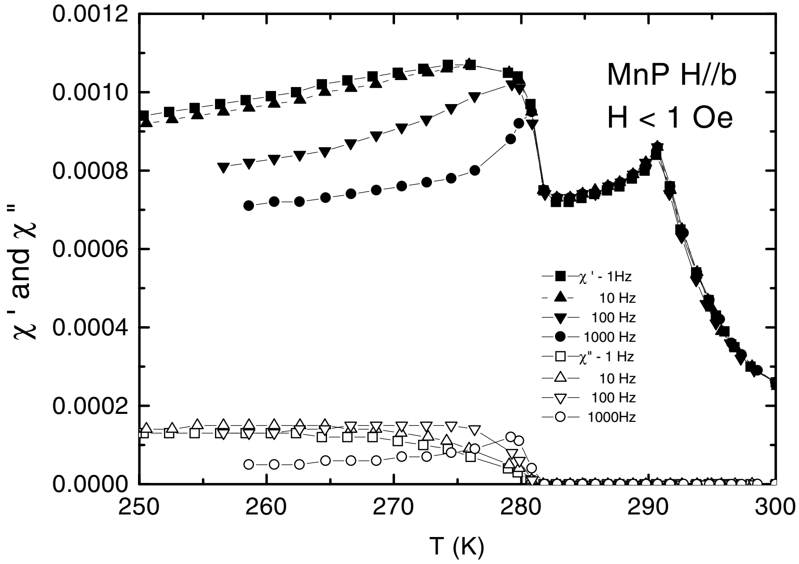


Figure 4. In-phase and out-of-phase components of the susceptibility against T in the region between 250 and 300 K for several values of the frequencies (1 Hz to 1 kHz). The applied field along the b axis is lower than 1 Oe.

over the region where the ferromagnetic phase is present although its value decreases as the screw transition is approached. At 60 K χ'' is already zero within the resolution of the instrument. For applied fields above 5 Oe (figure 3(b)) this transition is suppressed.

(3) The frequency dependences of $\chi'$and χ'' shown in figure 4 confirm the sharpness of the transition observed at 282 K since the sudden increase in χ' and χ'' is not frequency dependent. Within the new phase, the response of the system to higher frequency (f) decreases with increasing frequency.

The maximum at a temperature $T_m > T_C$ observed in the paramagnetic phase is due to the short-range order effects that usually accompany a second order ferromagnetic transition. These maxima, whose temperature T_m increases with increasing applied field, were previously observed in ferromagnetic insulators [8] and alloys [9] and more recently in $Tl_2Mn_2O_7$ [10]. This behaviour is explained in terms of the critical fluctuations that precede the ferromagnetic transition [10]. In MnP, they are observed in the $\chi'(T)$ curves at fixed H in the component of χ' along the easy and the intermediate anisotropy axis (b). In a ferromagnet a field applied along the easy axis destroys the transition and this manifests itself in the $\chi'(T)$ curves at constant H as a gradual suppression with H of the so-called 'Hopkinson peak' (see figure 1 of [11] for the phase diagram of MnP with H applied along the c axis), disclosing the maximum above T_C. However when a magnetic field $H_\perp$ is applied along a direction perpendicular to the easy axis, the phase transition preserves its characteristics and in the $H_\perp$–T plane a line of ordinary critical points of the same universality class as the one at $H = 0$ is generated [12]. The transition occurs now at a temperature $T_C(H)$ that decreases with increasing $H_\perp$. According to Riedel and Wegner [12] χ' at this second order transition diverges approaching this phase boundary either in temperature $(T - T_C(H))$ or in field $(H_\perp - H_c(T))$ with a lambda type anomaly that has a critical exponent related to the one of the specific heat. The susceptibility curves seen in figures 2 to 4 correspond to a field applied along the b axis (transverse to the ordering field that is along the c axis). When the transition occurs at $T_C(H)$ the cooperative response of the susceptibility increases abruptly. This is the second order transition marked by the characteristic lambda type anomaly that decreases in temperature with increasing H, disclosing, here also, the presence of the maximum of the susceptibility in the paramagnetic phase.

The sharp increases in χ' and χ'' at 282 K signal the occurrence of another phase transition. Since this increase occurs only in the susceptibilities along the $\boldsymbol{b}$ axis, it may be attributed to the appearance of a component of the magnetic moment along this axis. Either the onset of this component is continuous and then this will characterize the transition as second order or it will be abrupt and then the transition will be of first order. No thermal hysteresis was observed either at the transition at 292 K or at the one occurring at 282 K. In this range of temperatures with our experimental setup, we can only discriminate differences in temperatures between the cool-down and warm-up procedures if the temperatures differ by more than 0.2 K. The transition at 292 K is a well established second order transition where as can be seen in figures 3 and 4, no anomaly is observed in the out-of-phase component of the susceptibility. The sudden increase of the out-of-phase component of the susceptibility at 282 K may indicate that this new order–order transition can be of first order. The out-of-phase component of χ is related to the presence of a loss mechanism. If an order–order transition has a first order character a mixed phase may occur and domains of both phase may coexist. The oscillating AC field will cause a domain wall movement that can account for this absorption. Another possible explanation for the presence of this loss mechanism may be the occurrence of domains of the new phase along the $\boldsymbol{b}$ and $-\boldsymbol{b}$ directions. A domain wall movement with a corresponding absorption can also explain the onset of a non-zero χ''. In this last case a second order transition is compatible with the presence of a non-zero χ''.

The strong sensitivity of the susceptibility in this new phase to the small field, shown in figure 3(b), may indicate that a field of the order of 5 Oe is sufficient to saturate the component of the magnetization along the b axis. This is a similar situation to the one that occurs in weak ferromagnets where a very small field can completely suppress the peak observed in the transverse AC susceptibility at the transition [13]. A small applied field may also favour the suppression of domains along the $-\boldsymbol{b}$ direction and account for the absence of absorption above 5 Oe. If this is the case the transition at 282 K may be second order and the canting along the b direction may still be present for field above 5 Oe.

The decrease in the response (χ' and χ'') of this b component of the magnetization with increasing frequency (see figure 4) may occur because of its stiffness. As the frequency increases this component may not follow the oscillating field, causing the observed decrease in both components of χ.

The fact that this phase was not previously observed is probably due to the lack of precise measurements at very low fields in this region of temperature. It is worth noting that generally in experimental set-ups using superconducting or iron core electro-magnets the remanent field is higher than the 5 Oe needed to suppress this anomaly. We suggest that this phase transition is due to a weak spin reorientation of the moments at $T_r = 282$ K. As the temperature is lowered and approaches the ferro–screw transition the small angle that the ferromagnetic moments make with the c axis may decrease until they almost coincide with the c axis close to 47 K. Another plausible explanation would be that as the temperature is lowered the b component freezes causing a decrease in the response. In figure 1 the region corresponding to this new phase is indicated (not to scale).

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