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2002 J. Phys.: Condens. Matter 14 9779

(http://iopscience.iop.org/0953-8984/14/41/332)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **14** (2002) 9779–9784 PII: S0953-8984(02)36205-2

Interesting thermomagnetic history effects in the antiferromagnetic state of $SmMn_2Ge_2$

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Received 24 April 2002
Published 4 October 2002
Online at stacks.iop.org/JPhysCM/14/9779

Abstract
We present results of magnetization measurements showing that the magnetic response of the antiferromagnetic state of $SmMn_2Ge_2$ depends on the path used in the field (H)–temperature (T) phase space to reach this state. A distinct signature of metastability is observed in this antiferromagnetic state when obtained via field-cooling/field-warming paths.

The intermetallic compound $SmMn_2Ge_2$ with its interesting magnetic properties has been a subject of intensive study during last two decades [1–10]. In low applied magnetic fields it shows at least three magnetic transitions as a function of temperature [1–3, 6, 7]. First it undergoes a paramagnetic (PM)-to-ferromagnetic (FM1) transition at around 350 K, followed by an FM1-to-antiferromagnetic (AFM) transition at around 160 K (T_{N1}). On further reduction of the temperature, this AFM state transforms again into another ferromagnetic (FM2) state around 100 K (T_{N2}). There is a large spread in the reported values of the temperatures of the transitions from FM1 to AFM and from AFM to FM2 states. Quality of samples may be a possible source of the reported differences in transition temperatures, especially as it is known that the microscopic magnetic properties of RE (rare-earth) 1–2–2 compounds are quite sensitive to their underlying crystal lattice structure. On the other hand, there exists now enough evidence from various studies that both of these transitions are probably first order in nature [2, 4, 11, 12]. The first-order nature of these magnetic phase transitions can also provide a natural explanation for the reported spread in the transition temperatures. Supercooling (superheating) can take place down (up) to a temperature T^* (T^{**}) during cooling (heating) across a first-order transition point (T_N) [13]. The extent of supercooled/superheated phases will depend on the path followed in the field (H)–temperature (T) phase space [14]. In addition, in the samples with defect structures the lower- (higher-) temperature phase will start nucleating around these defect structures once the sample is cooled (heated) across T_N. This nucleation of the lower-T (higher-T) phase will be completed at T^* (T^{**}), and in the temperature regime T_N–T^* (T^{**}–T_N) there will be coexistence of two phases. All these properties will give rise

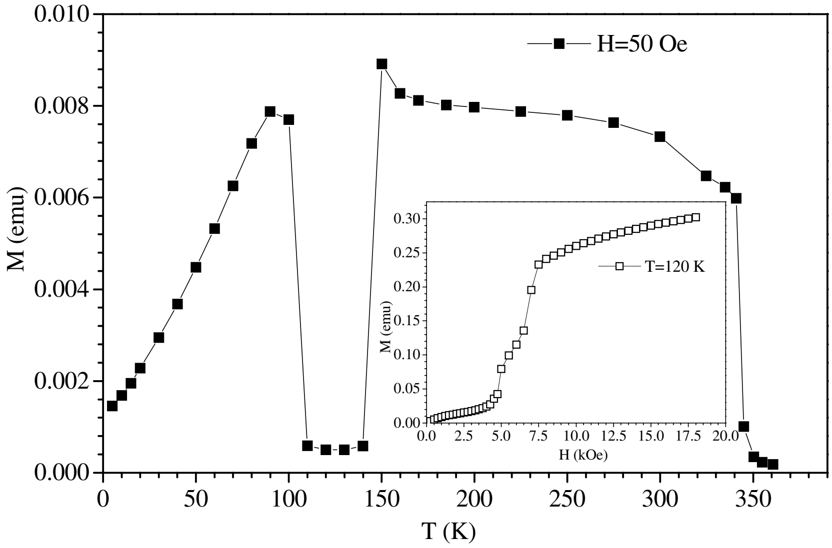

Figure 1. An M versus T plot for $SmMn_2Ge_2$ in an applied field of 50 Oe. The sample was ZFC to the lowest temperature before switching on the field, and data were taken while warming up the sample. The inset shows an M versus H curve plotted at $T = 120$ K after reaching that temperature under ZFC conditions.

to thermal hysteresis, and such thermal hysteresis is actually observed across the FM2–AFM and AFM–FM1 phase transitions in $SmMn_2Ge_2$ [2, 4, 7].

Confined between two FM phases at low and high temperatures and reached via first-order phase transitions, the AFM phase in $SmMn_2Ge_2$ is somewhat special. In this paper, on the basis of our careful dc magnetization studies we will show that the magnetic response of this AFM state actually depends on the path used in the (H, T) phase space to reach this state. A distinct trace of the high- (low-) temperature FM1 (FM2) state persists to well inside the AFM state, when this state is reached via a field-cooling (field-warming) path. We seek a possible explanation of the observed behaviour in terms of supercooling/superheating and phase coexistence across a first-order phase transition.

The $SmMn_2Ge_2$ sample used in our present study was prepared by argon arc melting and characterized by means of x-ray diffraction (XRD) measurements [6, 8]. The dc magnetization measurements were performed with a commercial SQUID magnetometer (MPMS-5, Quantum Design) using a 4 cm scan length.

Low-field ($H = 50$ Oe) magnetization (M) versus temperature (T) measurements reveal the FM1–AFM and AFM–FM2 transition temperatures for the present $SmMn_2Ge_2$ sample to be approximately 150 and 105 K respectively (see figure 1). We thus choose a temperature $T = 120$ K which is well inside the AFM state away from both the upper- and lower-temperature phase boundaries. Moreover, the M–H measurements at 120 K reveal a field-induced ferromagnetic transition around $H \approx 4$ kOe (see the inset of figure 1). To keep the sample well inside the AFM state in the (H, T) space, the applied field in the present measurements is limited to a maximum value of $H_{max} = 1$ kOe. We present in figure 2(a) the M–H plot at $T = 120$ K, measured after reaching this temperature under zero-field-cooled (ZFC) conditions. There is a small non-linearity as well as a small but distinct hysteresis in this M–H curve, which indicates that the magnetic state is not pure AFM in nature and there probably exists a finite amount of spin canting. The same feature may also arise from

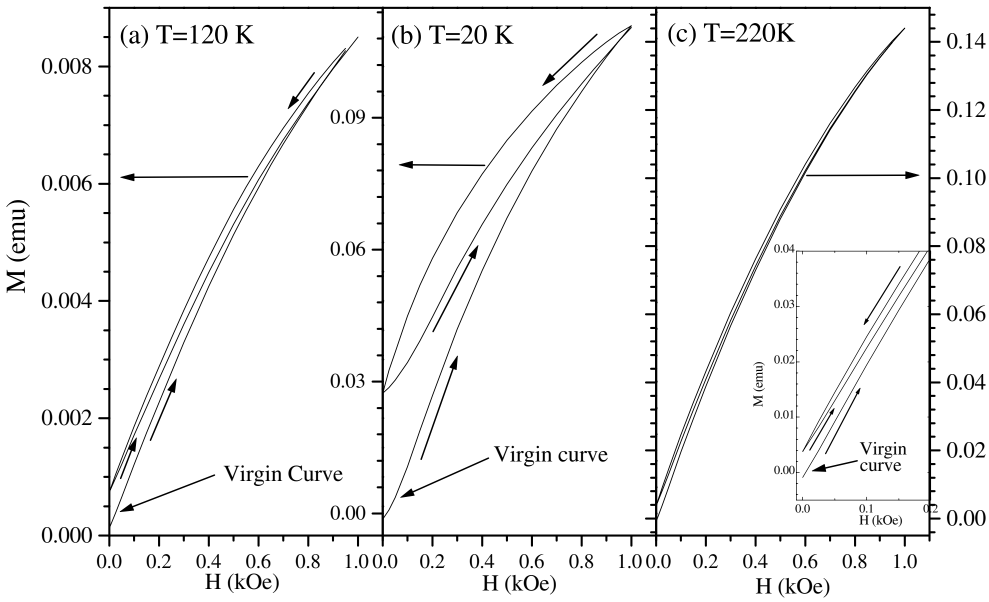

Figure 2. *M* versus *H* plots of $SmMn_2Ge_2$ obtained by field cycling between 0 and 1 kOe starting from ZFC conditions at (a) 120 K, (b) 20 K, and (c) 220 K. The field-cycling sequence is as follows: (1) the field is increased from 0 to 1 kOe; (2) it is decreased from 1 kOe to 0 Oe; (3) it is increased again from 0 to 1 kOe.

a very small amount (1–2%) of ferromagnetic phase which can go undetected in standard XRD measurements [6, 8]. In fact, similar non-linearity in the M–H curves in the AFM state of various $CeFe_2$-based pseudobinary alloys was earlier attributed to a small amount of ferromagnetic impurity phase which could not be detected in the XRD studies [15]. However, subsequent studies have indicated that this feature can be of intrinsic origin [16–18]. For our present system also, various studies (to be described below) indicate that the unconventional properties of the AFM state in $SmMn_2Ge_2$ cannot be explained in terms of a small amount of ferromagnetic impurity phase.

We shall now study the magnetic response of the AFM state at 120 K after reaching this state following three different experimental protocols:

(1) The temperature 120 K is reached from temperature well above T_{N1} in the absence of any applied field, and then a field of 1 kOe is switched on.
(2) A field of 1 kOe is switched on within the FM2 state at 4.5 K and the AFM state is reached subsequently by warming up the sample unidirectionally across T_{N2} to 120 K.
(3) A field of 1 kOe is switched on within the FM1 state at 300 K and the AFM state is reached subsequently by cooling the sample unidirectionally across T_{N1} to 120 K.

In all these experimental protocols, sufficient waiting time is allowed after reaching 120 K to ensure complete temperature stability. Furthermore, in experimental protocols Nos 1 and 3 the temperature is reduced slowly in small steps to avoid any temperature oscillation.

The values of the magnetization measured at 120 K with $H = 1$ kOe in both the protocols 2 and 3 are higher than that measured after switching on the field of 1 kOe at 120 K under the ZFC conditions, i.e. with protocol 1. This observation can be rationalized in terms of supercooling (superheating) of the FM1 (FM2) state. While cooling (warming) across the FM1 (FM2)–AFM transition temperatures T_{N1} (T_{N2}), some of the FM1 (FM2) state will supercool (superheat) into the temperature regime well beyond the transition temperature. The extent of the temperature regime of supercooling/superheating actually widens in the presence of the applied magnetic field [18, 19].

To test the above conjecture, we have studied the detailed nature of the magnetic state at $T = 120$ K obtained by the three different experimental protocols mentioned above. We use a '*minor-hysteresis-loop technique*' to show that the AFM state at 120 K obtained under ZFC conditions is a stable magnetic state, while the magnetic states obtained at 120 K by cooling/heating in the presence of a 1 kOe field are metastable in nature. Taking the M–H curve produced in the ZFC AFM state at 120 K by field cycling between 1 and 0 kOe as the envelope curve, we plot minor hysteresis loops (MHLs), terminating the M–H curve at various field points ($0 < H < 1$ kOe) on the descending-field leg of the envelope curve (see figures 2(a) and 3(a)). A distinct '*end-point memory*' is observed for all the MHLs: that is, on completion of the cycle, the MHLs show the same end-point magnetization value on the envelope M–H curve at 1 kOe. This kind of '*end-point memory*' is common to various kinds of hysteretic system including hard ferromagnets and superconductors [20, 21]. To emphasize this point we show in figures 2(b) and (c) similar M–H curves obtained by field cycling between 1 kOe and 0 Oe at $T = 20$ and 220 K which are well inside the FM2 and FM1 phases respectively. Clear '*end-point memory*' is observed in both cases.

We shall now describe the envelope curve and the MHLs in the AFM state at 120 K obtained by heating from 4.5 K in the presence of a field of 1 kOe, i.e. following protocol No 2. Compared with the ZFC case, the envelope curve and the MHLs are of very different nature (see figure 3(b)).The most prominent difference is the absence of '*end-point memory*'. Starting from the $H = 1$ kOe point and returning back to this point by producing MHLs of increasing field amplitude, a steadily decreasing value of magnetization is observed at the end point, i.e. $H = 1$ kOe. The end-point memory is in fact lost in the process of producing the first MHL by cycling between 1 and 0.8 kOe (see the paths marked 1 and 2 in the inset of figure 3(b)). The envelope curve produced by lowering the field from 1 kOe after this first cycle (see the path marked 3 in the inset of figure 3(b)) is quite different from the initial envelope curve. The differences in both the end-point magnetization and the envelope curves increase steadily with further cycling of the field with successively larger field amplitudes (see the paths marked 5, 6, 7, 8, 9, and 10 in the inset of figure 3(b)). The same field-cycling process does not have any effect on the end-point magnetization and envelope curve in the magnetic state obtained with experimental protocol No 1 (see figure 3(a)). This clearly shows that the initial magnetic state obtained with protocol No 2 is a metastable state, and the energy fluctuations introduced while producing the MHLs steadily push this state towards the stable ZFC state. This kind of lack of '*end-point memory*' was earlier observed across the vortex matter phase transition from one kind of vortex solid to another in a type-II superconductor $CeRu_2$ (see figure 7 of [22]). This was attributed to the existence of metastable states across a disorder-broadened first-order transition [22]. These metastable states were shattered by the energy fluctuations generated while producing the MHLs in the (H, T) regime concerned. The same behaviour was observed subsequently across a disorder-induced first-order transition in the vortex matter phase space of another type-II superconductor, $NbSe_2$ [23].

The same lack of *'end-point memory'* effect is observed at 120 K on cooling from 300 K to well inside the FM1 state, i.e. following protocol No 3 (see figure 3(c)). On the other hand, experiments of the same kind, carried out after preparing the magnetic states well inside the ferromagnetic regions FM1 and FM2 by crossing the transition temperatures T_{N1} and T_{N2} in the presence of a field, show clear *'end-point memory'* effects (data not shown here for the sake of clarity and conciseness). These results also rule out any possible contribution

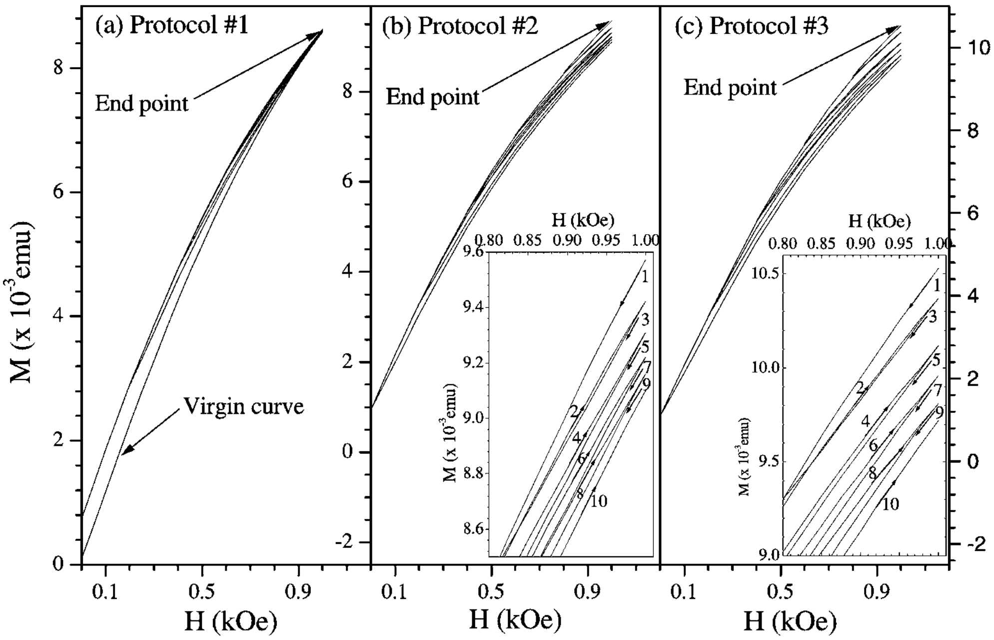

Figure 3. M versus H plots and MHLs with field cycling between 0 and 1 kOe for $SmMn_2Ge_2$ at 120 K obtained with three different experimental protocols (see the text for details). (a) Results obtained with protocol No 1. (b) Results obtained with protocol No 2. The inset shows the MHLs plotted for the following field sequence: H is decreased from 1 to 0.8 kOe (path 1), increased from 0.8 Oe back to 1 kOe (path 2), decreased from 1 to 0.6 kOe (path 3), increased from 0.6 to 1 kOe (path 4), decreased from 1 to 0.4 kOe (path 5), increased from 0.4 to 1 kOe (path 6), decreased from 1 to 0.2 kOe (path 7), increased from 0.2 to 1 kOe (path 8), decreased from 1 kOe to 0 Oe (path 9), and lastly increased from 0 Oe to 1 kOe (path 10). The last two sequences essentially form the envelope curve. Note that the end-point magnetization at $H = 1$ kOe decreases progressively. The same set of sequences is followed for producing MHLs in (a). In contrast to the present case, the end-point magnetization at $H = 1$ kOe retains the same value, confirming the end-point memory. (c) Results obtained with protocol No 3. The inset shows the MHLs plotted for the following field sequence: H is decreased from 1 to 0.8 kOe (path 1), increased from 0.8 Oe back to 1 kOe (path 2), decreased from 1 to 0.6 kOe (path 3), increased from 0.6 to 1 kOe (path 4), decreased from 1 to 0.4 kOe (path 5), increased from 0.4 to 1 kOe (path 6), decreased from 1 to 0.2 kOe (path 7), increased from 0.2 to 1 kOe (path 8), decreased from 1 kOe to 0 Oe (path 9), and lastly increased from 0 Oe to 1 kOe (path 10). The last two sequences essentially form the envelope curve. Note that the end-point magnetization at $H = 1$ kOe decreases progressively as in (b), showing a lack of end-point memory.

from an impurity ferromagnetic phase in the observed metastable behaviour of the AFM state. Metastability (if any) related to the hindrance of domain motions in a ferromagnet is known to be greater while warming up from the low-temperature ZFC state than while cooling down from the high-temperature region in the presence of an applied field [24].

This lack of '*end-point memory*' is now accepted as a signature of metastability associated with a first-order transition [22, 23], which is taken as support for the proposed first-order nature of certain vortex matter phase transitions in type-II superconductors [22, 23, 25]. In this paper we have looked at the AFM state of $SmMn_2Ge_2$, which can be reached from both high- and low-temperature FM states through magnetic phase transitions. The first-order nature of these two transitions has already been considered [2, 4, 11, 12]. We have now shown the lack of '*end-point memory*' in the MHLs and associated metastable behaviour in this AFM state for

$SmMn_2Ge_2$. The observed behaviour highlights the interesting status of the AFM state in $SmMn_2Ge_2$ sandwiched between two FM states in the (H, T) phase space and reached via first-order phase transitions.

Summarizing our results, we find interesting thermomagnetic history effects well inside the AFM state of $SmMn_2Ge_2$. When this AFM state is reached from the FM1 (FM2) state by cooling (heating) in the presence of an applied field, the traces of the FM1 (FM2) state remain as a supercooled (superheated) state. This is an example of phase coexistence of the AFM–FM1 (FM2) state, and the resulting magnetic state is metastable in nature. In producing MHLs in this metastable state one introduces energy fluctuations, which drive the domains of the metastable FM1 (FM2) state to the stable AFM state. Such metastability in the form of a lack of '*end-point memory*' (which is also observed across solid-to-solid vortex matter phase transitions in various type-II superconductors [22, 23]) may serve as a characteristic signature of a first-order transition in samples with substantial defect structures where the detection of latent heat as the canonical signature of a first-order transition is relatively difficult [26].

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