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

S. B. Roy[1], S. Chaudhary[1], M. K. Chattopadhyay [1], P. Chaddah[1] and E. V. Sampathkumaran[2]

[1] *Low Temperature Physics Laboratory, Centre for Advanced Technology, Indore 452013, India*

[2] *Tata Institute of Fundamental Research, Mumbai 400005, India*

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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. Distinct signature of metastablity is observed in this antiferromagnetic state when obtained via field-cooling/field-warming paths. The isothermal M-H loops show lack of end-point memory, reminiscent of that seen in metastable vortex states near the field-induced first order phase transition in various type-II superconductors.

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 350K, followed by an FM1 to antiferromagnetic (AFM) transition at around 160 K (T_{N1}). On reduction of the temperature further 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 transition temperatures from FM1 to AFM and AFM to FM2 states. Quality of the samples may be a possible source for the reported differences in the transition temperatures, especially when it is known that the microscopic magnetic properties of RE(rare-earth)1-2-2 compomuds are quite sensitive to their underlying crystal lattice structure. On the other hand, there exist now enough evidences 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 to the reported spread in the transition temperatures. Supercooling(superheating) can take place down(up) to a temperature T^* (T^{**}) while 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(higher) T phase will be completed at T^*(T^{**}), and in the temperature regime T_N-T^* (T^{**}-T_N) there will be co-existence of two phases. All these properties will give rise 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 phase at low and high temperature and reached via first order phase transitions, the AFM phase in $SmMn_2Ge_2$ is something special. In this paper, based on our careful dc magnetization studies we shall 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. Distinct trace of the high(low) temperature FM1(FM2) state remains well inside the AFM state, when this state is reached via a field cooling(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 X-ray diffraction(XRD) measurements [6,8]. Dc magnetization measurements were performed with a commercial SQUID-magnetometer (MPMS-5, Quantum Design) using a 4 cm scan length.

A low field (H=50 Oe) magnetization(M) versus temperature(T) measurement reveal the FM1-AFM and AFM-FM2 transition temperatures in the present $SmMn_2Ge_2$ sample to be approximately 150 K and 105 K respectively (see Fig.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 120K reveal a field-induced ferromagnetic transition around H≈4 kOe (see the inset of Fig.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 Fig.2(a) the M-H plot at T=120 K, measured after reaching this temperature in zero field cooled (ZFC) condition. 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 antiferromagnetic in nature and there probably exists a finite amount of spin-canting. The same feature may also arise from 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 antiferromagnetic state of various $CeFe_2$ based pseudobinary alloys, has earlier been attributed to 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]. In our present system also various studies (to be narrated below) indicate that the unconventional properties of the antiferromagnetic 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 120K after reaching this state following the three different experimental protocols :

1. the temperature 120 K is reached from temperature well above T_{N1} in 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 4.5K and the AFM state is reached subsequently by warming up the sample unidirectionally across T_{N2} to 120K.

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 120K.

In all these experimental protocols sufficient wait time is given after reaching 120K to ensure complete temperature stability. Furthermore, in experimental protocol no. 1 and 3 temperature is reduced slowly in small steps to avoid any temperature oscillation.

The values of 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 120K in the ZFC condition i.e.with the protocol 1. This observation can be rationalized in terms of supercooling (superheating) of the FM1(FM2) state. While cooling(warming) acrosss the FM1(FM2)-AFM transition temperatures T_{N1}(T_{N2}) some amount of the FM1(FM2) state will supercool (superheat) into the temperature regime well beyond the transition temperature. The extent of temperature regime of supercooling/superheating actually widens in presence of the applied magnetic field [18,19].

To support the above conjecture we have studied the detailed nature of the magnetic state at T=120K obtained by three different experimental protocols mentioned above. We use a "*minor hysteresis loop technique*" to show that the AFM state at 120K obtained in ZFC condition is a stable magnetic state, while the magnetic states obtained at 120K by cooling/heating in presence of 1 kOe field are metastable in nature. Taking the M-H curve drawn in the ZFC AFM state at 120K by field cycling between 1 kOe and 0 kOe as the envelope curve, we draw minor hysteresis loops (MHLs), terminating the M-H curve at various field points ($0 < H < 1kOe$) on the the descending field leg of the envelope curve (see Fig. 2(a) and 3(a)). A distinct "*end point memory*" is observed for all the MHls, namely 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 with various kinds of hysteretic systems including hard ferromagnets and superconductors [20,21]. To emphasise this point we show in Fig. 2 (b) and 2 (c) similar M-H curves obtained by field cycling between 1 k Oe and 0 Oe at T=20K and 220 K which are well inside the FM2 and FM1 phase respectively . Clear "*end point memory*" is observed in both the cases.

We shall now draw the envelope curve and the MHLs in the AFM state at 120K obtained by heating from 4.5 K in the presence of a field of 1 kOe i.e. following the protocol no.2. In contrast to the ZFC case, the envelope curve and the MHLs are of very different nature (see Fig.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 drawing 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 drawing the first MHL by cycling between 1 kOe and 0.8 kOe (see the paths marked by 1 and 2 in the inset of fig. 3(b)). The envelope curve drawn by lowering the field from 1 kOe after this first cycle (see path marked by 3 in the inset of Fig.3) is quite different from the initial envelope curve. The differences in both the end point magnetization and the envelope curves rise steadily with further cycling of field with succesively larger field amplitude (see path marked by 5,6,7,8,9 and 10 in the inset of Fig.3(b)). Same field cycling process does not cause any effect on the end point magnetization and envelope curve in the magnetic state obtained with the experimental protocol no.1 (see Fig. 3(a)). This clearly shows that the initial magnetic state obtained with protocol no. 2 is a metastable state, and the energy fluctuation introduced while drawing the MHLs steadily push this state towards the stable ZFC state. This kind of the lack of "*end point memory*" has earlier been observed across the vortex matter phase transition from one kind of vortex solid to another in a type-II superconductor $CeRu_2$ (see Fig. 7 of Ref.22). This was attributed to the existence of metastable states across a disorder broadened first order transition [22]. These metastable states were shattered through the energy fluctuation generated while drawing the MHLs in the concerned (H,T) regime. The same behaviour has been 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 form 300 K well inside the FM1 state i.e. following the protocol no.3 (see Fig. 3(c)). On the other hand same kind of experiments after preparing the magnetic states well inside the ferromagnetic regions FM1 and FM2 by crossing the transition temperatures T_{N1} and T_{N2} in presence of field, show clear "*end point memory*" effect (data not shown here for the sake of clarity and conciseness). These results also negate any possible contribution 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 more 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 a support for establishing the 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 state through magnetic phase transitions. The first order nature of these two transitions is already considered [2,4,11,12]. We have now shown the lack of "*end point memory*" in the minor hysteresis loops and associated metastable behaviour in this AFM state of $SmMn_2Ge_2$. 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.

Summarising our results, we find interesting thermomagnetic history effects well inside the AFM state of $SmMn_2Ge_2$. When this AFM state is reached either from the FM1(FM2) state by cooling (heating) in presence of an applied field, the traces of the FM1(FM2) state remain as 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. On drawing MHLs in this metastable state one introduces energy fluctuations, which drive the domains of metastable FM1(FM2) state to the stable AFM state. Such metastability in the form of the lack of "*end point memory*" (which is also observed across solid to solid vortex matter phase transitions in various type-II superconductors (ref.22 and 23)) may serve as characteristic signatures 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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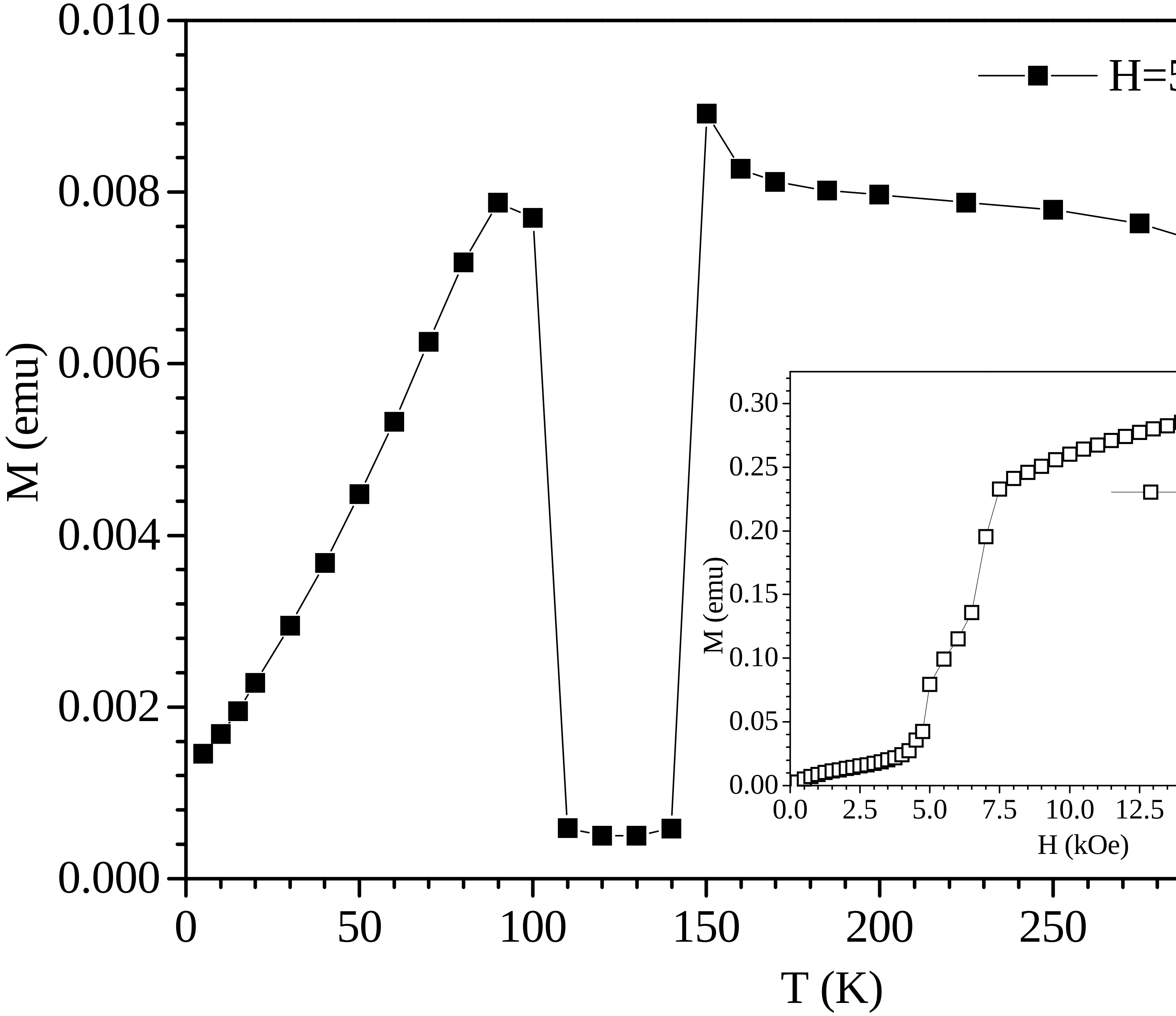
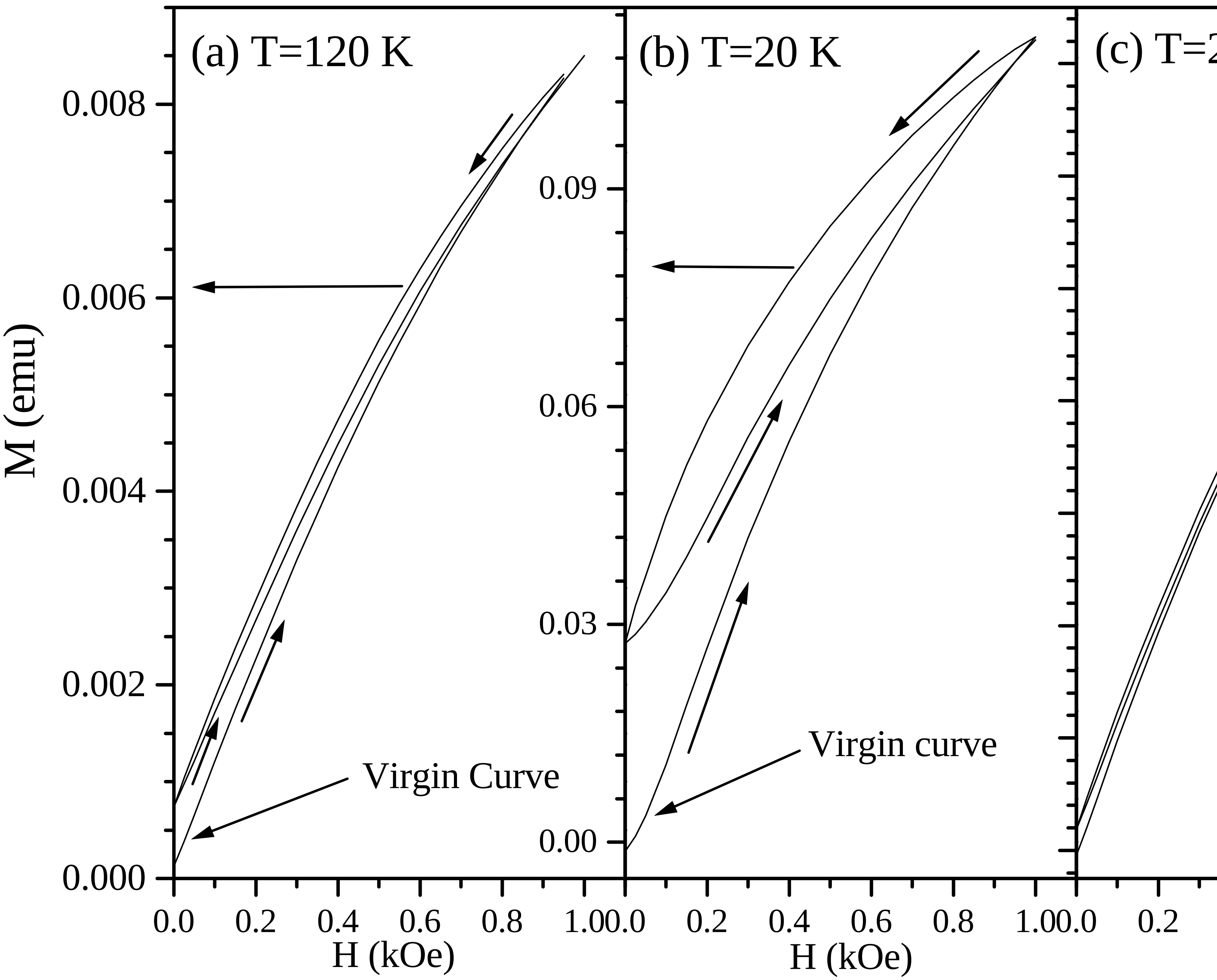
FIGURES

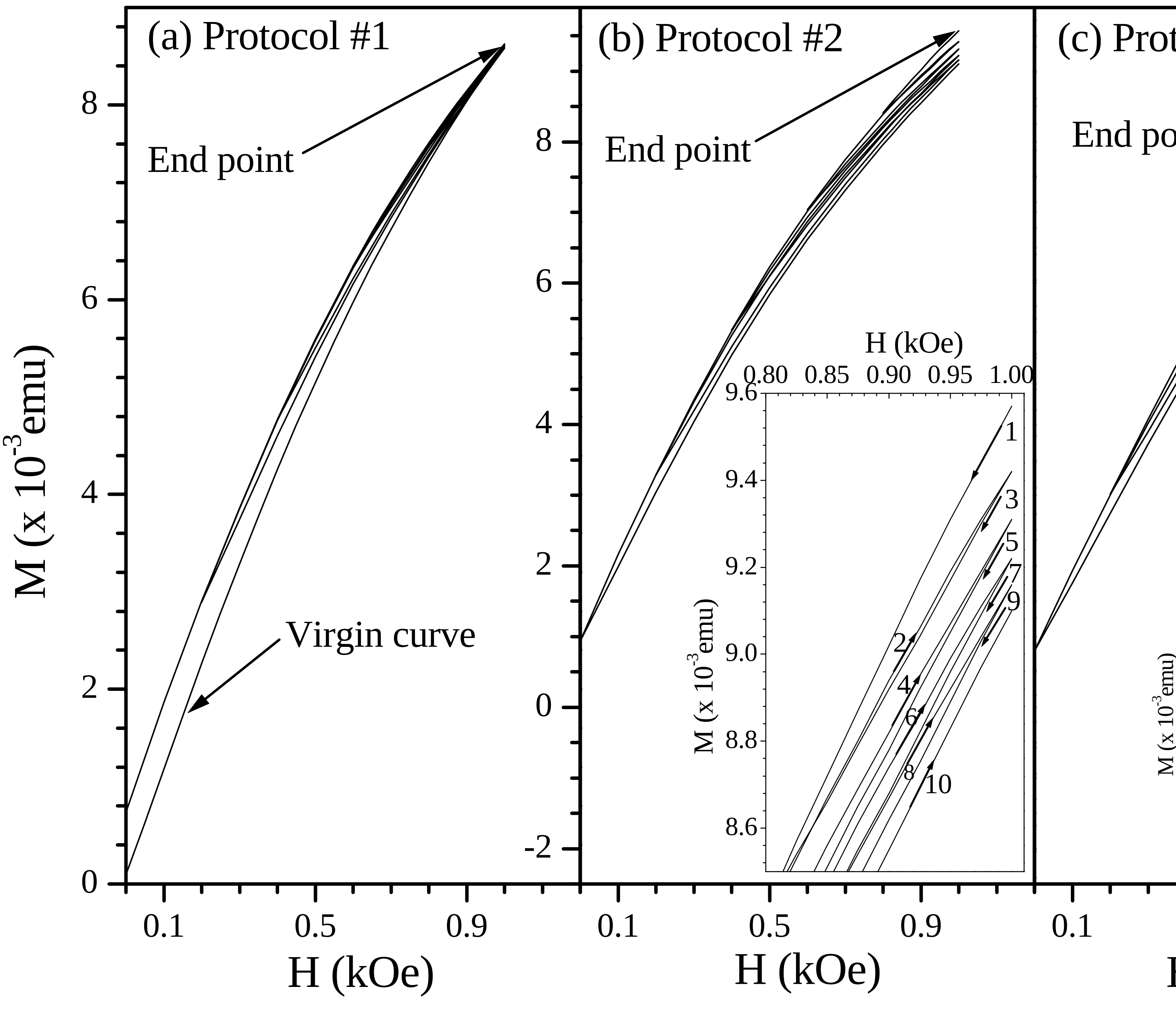
FIG. 1. M vs T plot of $SmMn_2Ge_2$ in an applied field of 50 Oe. The sample was zero field cooled to the lowest temperature before switching on the field, and data was taken while warming up the sample. The inset shows M vs H curve drawn at T=120K after reaching that temperature in zero field cooled condition.

FIG. 2. M vs H plot of $SmMn_2Ge_2$ obtained by field cycling between 0 and 1 kOe starting from zero field cooled condition at (a) 120 K, (b) 20 K and (c) 220K. The field cycling sequence is the following: (1) the field is increased from 0 to 1 kOe, (2) decreased from 1 kOe to 0 Oe (3) increased again from 0 to 1 kOe.

FIG. 3. M vs H plot and minor hysteresis loops with field cycling between 0 and 1 kOe of $SmMn_2Ge_2$ at 120 K obtained with three different experimental protocols (see text for details). (a) Results obtained with protocol no. 1. (b) Results obtained with protocol no. 2. The inset shows the minor hysteresis loops drawn in the following field sequence. H is decreased from 1 kOe to 0.8 kOe (path 1), increased from 0.8 Oe back to 1 kOe (path 2), decreased from 1 kOe to 0.6 kOe (path 3), increased from 0.6 kOe to 1 kOe (path 4), decreased from 1 kOe to 0.4 kOe (path 5), increased from 0.4 kOe to 1 kOe (path 6), decreased from 1 kOe to 0.2 kOe (path 7), increased from 0.2 kOe 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 sequence essentially forms the envelope curve. Note that the end point magnetization at H=1 kOe decreases progressively. The same set of sequence is followed for drawing minor hysteresis loops fig. 3(a). In contrast to the present case the end point magnetization at H=1 kOe retains the same value confirming end point memory. (c) Results obtained with protocol no. 3. The inset shows the minor hysteresis loops drawn in the following field sequence. H is decreased from 1 kOe to 0.8 kOe (path 1), increased from 0.8 Oe back to 1 kOe (path 2), decreased from 1 kOe to 0.6 kOe (path 3), increased from 0.6 kOe to 1 kOe (path 4), decreased from 1 kOe to 0.4 kOe (path 5), increased from 0.4 kOe to 1 kOe (path 6), decreased from 1 kOe to 0.2 kOe (path 7), increased from 0.2 kOe 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 sequence essentially forms the envelope curve. Note that the end point magnetization at H=1 kOe decreases progressively as in fig. 3(b), showing lack of end point memory.


0.010
0.008
0.006
0.004
0.002
0.000
M (emu)
0
50
100
150
200
250
T (K)
H=5
0.30
0.25
0.20
0.15
0.10
0.05
0.00
M (emu)
0.0
2.5
5.0
7.5
10.0
12.5
H (kOe)


(a) T=120 K
(b) T=20 K
(c) T=2
M (emu)
H (kOe)
H (kOe)
Virgin Curve
Virgin curve
0.000
0.002
0.004
0.006
0.008
0.00
0.03
0.06
0.09
0.0
0.2
0.4
0.6
0.8
1.0



(a) Protocol #1
End point
Virgin curve
M (x 10^{-3}emu)
H (kOe)
(b) Protocol #2
End point
H (kOe)
0.80 0.85 0.90 0.95 1.00
9.6
9.4
9.2
9.0
8.8
8.6
M (x 10^{-3}emu)
1
2
3
4
5
6
7
8
9
10
H (kOe)
(c) Prot
End po
M (x 10^{-3}emu)