Pressure-induced changes in the magnetic and magnetocaloric properties of $R\mathrm{Mn_2Ge_2}$ (R=Sm,Gd)

Pramod Kumar,[1] K. G. Suresh,[1,*] A. K. Nigam,[2] A. Magnus,[3] A. A. Coelho,[3] and S. Gama[3]
[1]*Department of Physics, Indian Institute of Technology Bombay, Mumbai 400076, India*
[2]*Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai 400005, India*
[3]*Instituto de Física 'Gleb Wataghin,' Universidade Estadual de Campinas-UNICAMP, CP 6165, Campinas 13 083 970, SP, Brazil*
(Received 17 December 2007; revised manuscript received 15 April 2008; published 17 June 2008)

We have studied the variation of magnetic and magnetocaloric properties of polycrystalline compounds $\mathrm{SmMn_2Ge_2}$ and $\mathrm{GdMn_2Ge_2}$ as a function of applied hydrostatic pressure. The magnetic transition temperatures are found to change considerably with pressure. The temperature regime of existence of antiferromagnetic (AFM) ordering is found to increase with pressure, in both the compounds. In $\mathrm{SmMn_2Ge_2}$, the sign of the magnetocaloric effect at the low-temperature ferromagnetic (FM)-AFM transition changes with pressure. The isothermal magnetic entropy change in this compound is found to increase by about 20 times as the pressure is increased from the ambient value to 6.8 kbar. Effect of pressure in $\mathrm{GdMn_2Ge_2}$ is less compared to that in $\mathrm{SmMn_2Ge_2}$. The variations in the magnetic and magnetocaloric properties are attributed to the changes in the magnetic state of the Mn sublattice under pressure. The difference in R-Mn coupling in Sm and Gd compounds is also found to play a role in determining the magnetic and magnetocaloric properties, both at ambient as well as under applied pressures.

DOI: 10.1103/PhysRevB.77.224427 PACS number(s): 75.30.Sg, 71.20.Eh, 75.30.Kz

I. INTRODUCTION

The study of magnetocaloric effect (MCE) has become an important area of research in the field of magnetic materials owing to the possibility of developing some of them into potential magnetic refrigerants.[1–3] MCE is usually measured either in terms of isothermal magnetic entropy change (ΔS_M) or adiabatic temperature change (ΔT_{ad}). Materials with first-order magnetic transition are of particular interest since they exhibit significant MCE at the magnetic transition temperature. Compounds showing field-induced magnetic transitions and/or structural transitions have been found to exhibit giant MCE, as in the case of $\mathrm{Gd_5(Si_{1-x}Ge_x)_4}$.[3–5] Among the rare earth (R)-transition metal intermetallic compounds, $R\mathrm{Mn_2Ge_2}$ compounds, in general, display interesting magnetic properties such as multiple magnetic transitions, first-order transitions, and re-entrant ferromagnetism.[6–10] Another important aspect of this class of compounds is the strong magnetovolume effect. Among the various compounds of this series, $\mathrm{SmMn_2Ge_2}$ has attracted special attention owing to the anomalous magnetic and related properties.[10–13] As a probe to understand the magnetovolume effect, we have studied the effect of hydrostatic pressure on the magnetic properties of $\mathrm{SmMn_2Ge_2}$. The same studies have also been carried out on $\mathrm{GdMn_2Ge_2}$ which is isostructural to $\mathrm{SmMn_2Ge_2}$, but which shows quite different magnetic properties. While the crystal-field effect is expected to be considerable in $\mathrm{SmMn_2Ge_2}$, it should be practically absent in $\mathrm{GdMn_2Ge_2}$ due to the S-state nature of the $4f$ shell of $\mathrm{Gd^{3+}}$ ion. It should also be kept in mind that while the Sm-Mn coupling is ferromagnetic in nature, the Gd-Mn coupling is antiferromagnetic (AFM), due to the fact that Sm is a light rare earth and Gd is a heavy rare earth.

The anomalous magnetic properties of $\mathrm{SmMn_2Ge_2}$ include multiple magnetic transitions, re-entrant ferromagnetism, large positive magnetoresistance in the reentrant FM phase etc. Chaudhary *et al.*[14] have reported that while in low fields the field cooled (FC) magnetization in the re-entrant FM phase is larger than that of the high temperature FM phase, the trend reverses at high fields. Another observation is that the FC magnetization becomes less than the zero-field cooled (ZFC) magnetization in the re-entrant phase, as the field is increased above a critical value. Though a reasonable understanding is achieved for some of the above observations, there seem to exist some questions which remain unanswered even today.

$\mathrm{SmMn_2Ge_2}$ is known to possess three magnetically ordered phases, namely the ferromagnetic ($146\ \mathrm{K}<T<350\ \mathrm{K}$), antiferromagnetic ($110\ \mathrm{K}<T<146\ \mathrm{K}$), and re-entrant ferromagnetic ($T<110\ \mathrm{K}$) phases. The transitions at $T_1=110\ \mathrm{K}$ and $T_2=146\ \mathrm{K}$ are known to be first order in nature. It has been reported that the magnetic properties of this compound mainly arises from the strong structure-magnetic property dependence. On the other hand, $\mathrm{GdMn_2Ge_2}$ shows only one magnetic transition, i.e., at 95 K, below the room temperature. The main difference in the magnetic properties of these two compounds is attributed to the difference in their unit-cell parameters and the Mn-Mn bond length. In view of this, it is of great interest to compare the magnetic properties of these two compounds as a function of applied pressure. As magnetocaloric effect is closely related to the magnetic state of the material, it is of added importance to investigate the variation of magnetocaloric effect as a function of applied pressure. Though some reports are available on the magnetic properties of some $R\mathrm{Mn_2Ge_2}$ compounds under pressure,[15,16] to the best of our knowledge, only one report[17] is available on the pressure dependence of MCE.

II. EXPERIMENTAL DETAILS

The preparation and the characterization techniques for $\mathrm{SmMn_2Ge_2}$ and $\mathrm{GdMn_2Ge_2}$ have been reported

 ©2008 The American Physical Society

elsewhere.[18,19] The magnetization (M) measurements under various applied pressures (P) have been performed using in a superconducting quantum interference device (SQUID) magnetometer (Quantum Design) attached with a Cu–Be clamp type pressure cell (maximum pressure ~12 kbar). The magnetic transition temperatures have been calculated from the (dM/dT) plots obtained from the ZFC M-T data. In this mode, the samples were cooled in zero field (H) and the magnetization was measured under a field of 200 Oe in the warming cycle. The MCE of all the compounds have been determined in terms of the isothermal magnetic entropy change using the M-H-T data collected close to the transition temperatures. Throughout the text, $P=0$ implies the ambient pressure.

III. RESULTS

The Rietveld refinement of room-temperature powder x-ray diffractograms confirms that both $SmMn_2Ge_2$ and $GdMn_2Ge_2$ have formed in single phase with the $ThCr_2Si_2$ tetragonal structure (space group=$I4/mmm$). The lattice parameter were found to be $a=4.062$ Å and $c=10.889$ Å for $SmMn_2Ge_2$. The corresponding values of $GdMn_2Ge_2$ are 4.026 and 10.881 Å. The intralayer ($d^a_{\text{Mn-Mn}}$) and interlayer ($d^c_{\text{Mn-Mn}}$) distances are 2.872 and 5.445 Å, respectively, for $SmMn_2Ge_2$, while they are 2.847 and 5.441 Å for $GdMn_2Ge_2$.[18,19] According to a model [interlayer coupling (ITC)] proposed by Fujiwara *et al.*,[20] if $d^a_{\text{Mn-Mn}}>2.85$ Å, then the interlayer exchange coupling constant ($J^c_{\text{Mn-Mn}}$) is positive (i.e., FM). For $d^a_{\text{Mn-Mn}}<2.85$ Å, $J^c_{\text{Mn-Mn}}$ is negative (AFM). Another model [intralayer coupling (IRC)] proposed by Welter *et al.*[21] has shown that the value of $d^a_{\text{Mn-Mn}}$ not only affects $J^c_{\text{Mn-Mn}}$, but also the intralayer Mn-Mn exchange, namely $J^a_{\text{Mn-Mn}}$. There exists a critical distance of 2.84 Å above which the intralayer Mn-Mn coupling is noncollinear ferromagnetic and below which it is collinear ferromagnetic. The noncollinear ferromagnetic coupling leads to canting of the Mn moments in the ab plane. From the $d^a_{\text{Mn-Mn}}$ values listed above, it can be seen that in $SmMn_2Ge_2$ at room temperature, the value is more than the critical values, according to both the models. Koyama *et al.*[12] have reported that there is an abrupt change in the unit-cell volume as the materials enter the AFM phase on cooling. These authors have also shown that in presence of a field, there is no abrupt change in the lattice parameters near T_1 as well as T_2. On the other hand, in $GdMn_2Ge_2$, $d^a_{\text{Mn-Mn}}$ value is just below the critical distance as per the ITC model and just above the critical distance as per the IRC model.

According to the above two spacing conditions, $SmMn_2Ge_2$ shows a canted FM state below the ordering temperature (T_C) of 350 K. On cooling, due to the thermal contraction of the lattice parameters and the bond lengths, it undergoes a transition to a noncollinear AFM at $T_2=146$ K. The AFM phase is retained down to 110 K. On further cooling, the magnetic ordering of the Sm sublattice causes sufficiently large molecular field and the compound attains a reentrant canted FM phase below $T_1=110$ K. In addition, as reported by Chaudhary *et al.*,[14] another transition at about 35 K is also observed. However, this transition could not be observed from heat capacity, NMR, ac susceptibility or neutron-diffraction measurements, and the exact origin of this transition is not yet well understood.

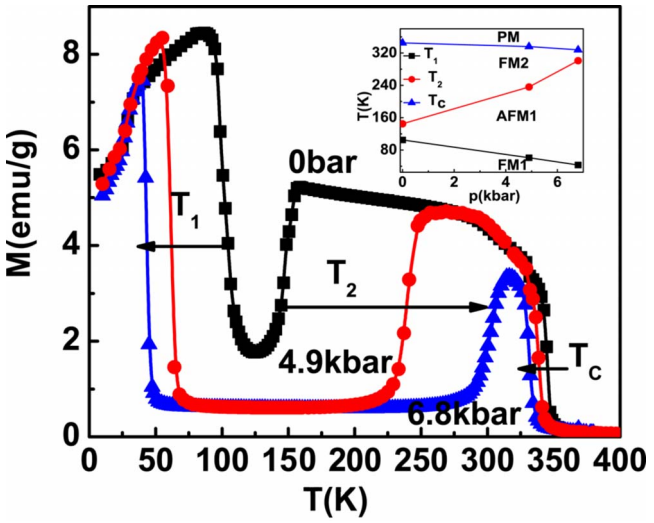

FIG. 1. (Color online) Temperature dependence of magnetization of $SmMn_2Ge_2$ in a magnetic field of 200 Oe under different applied pressures. The inset shows the variation of transition temperatures T_1, T_2, and T_C with pressure. FM1 refers to the re-entrant ferromagnetic phase, FM2 refers to the high-temperature ferromagnetic phase, and PM refers to the paramagnetic state.

Figure 1 shows the temperature dependence of the ZFC magnetization data of $SmMn_2Ge_2$ obtained under different pressures in an applied field of 200 Oe. It is evident from the figure that with the application of pressure, the transitions at T_1 and T_C shift to low temperatures, whereas the one at T_2 shifts to high temperatures. Inset of Fig. 1 shows the variation of T_1, T_2, and T_C with pressure. Both T_1 and T_C decrease almost linearly with pressure, at the rates of 9.1 and 2.3 K/kbar, respectively. The rate of increase of T_2 is found to be 22.3 K/kbar. With increase in pressure, the temperature range of the existence of AFM ordering has increased considerably. Furthermore, the magnetization in the AFM region has decreased considerably with pressure. These observations can be attributed to the reduction in the lattice parameters and the Mn-Mn bond length, which leads to an increase in the AFM component in the Mn sublattice. Tomka *et al.* have also observed the same behavior in $SmMn_2Ge_2$.[15] It is to be noted that the transition seen at about 35 K at ambient pressure remains unaltered with pressure. This transition must be purely associated with the Sm sublattice as revealed by its insensitiveness to pressure.

Recently we have reported[19] the effect of chemical pressure brought about by Si substitution for Ge in $SmMn_2Ge_2$. It was found that the transition temperature T_1 decreases from 110 to 39 K, as Ge is completely replaced with Si. It was also observed that Si substitution has caused a decrease in the lattice parameters, which implies that there is a chemical pressure as a result of Si substitution. Therefore, the present observation of reduction in T_1 is consistent with the substitutional studies. In both the cases, a decrease in the Mn-Mn intra- and interlayer distances causes a reduction in the ferromagnetic Sm-Mn exchange and hence T_1 decreases. A

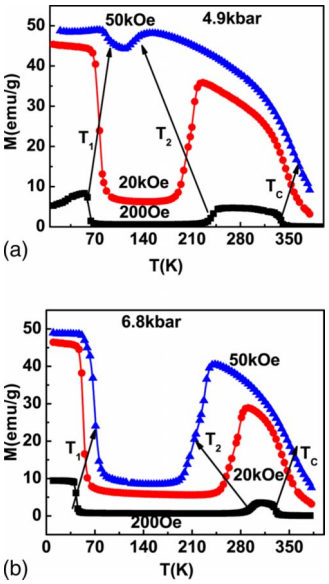

FIG. 2. (Color online) Temperature dependence of magnetization of $SmMn_2Ge_2$ in different fields under applied pressures of (a) 4.9 kbar and (b) 6.8 kbar.

similar observation has been reported in $La_{0.75}Sm_{0.25}Mn_2Si_2$ by Gerasimov *et al.*[16]

Figures 2(a) and 2(b) show the M-T plots obtained in $SmMn_2Ge_2$ under fields of 200 Oe, 20 kOe, and 50 kOe for applied pressures of 4.9 and 6.8 kbar, respectively. At ambient as well as at high pressures, the transition temperature T_1 and T_C are found to increase while T_2 is found to decrease with field. These observations confirm that the magnetic phases just below T_1 and T_C are FM, while that below T_2 is AFM.[16] For both 4.9 and 6.8 kbar, the AFM regime is found to narrow down with increase in field. It is of interest to note that the AFM region is considerably suppressed by 50 kOe, when the pressure is 4.9 kbar. When the pressure is increased to 6.8 kbar, the AFM region extends over a longer temperature range for the same field (50 kOe). Therefore, the variations seen in Figs. 2(a) and 2(b) clearly imply that the strength (and the temperature range of existence) of the AFM ordering is determined by the competition between the pressure and the magnetic field and that these two factors contribute almost equally at 4.9 kbar and at 50 kOe. This is consistent with the observation reported by Chaudhary *et al.*,[14] which shows that there is no dip (due to the formation of AFM state) in the magnetization at ambient pressures for fields of 20 and 50 kOe. Koyama *et al.*[13] and Han *et al.*[14] have also reported that at ambient pressure, the AFM state is suppressed by the application of a field of about 10 kOe. In contrast, the M-T plots obtained in 6.8 kbar and in fields of 20 and 50kOe [Fig. 2(b))] show that the transition at T_1 is very sharp and that the AFM region is quite prominent. These results suggest that the AFM phase gets stabilized by the application of pressure and that a pressure of 6.8 kbar is sufficient enough to retain the AFM phase ($T_1 < T < T_2$) even when the applied field is as high as 50 kOe.

The field dependence of magnetization isotherms obtained at 10 K of $SmMn_2Ge_2$ for different pressures are found to show a FM behavior with rapid increase in magnetization at

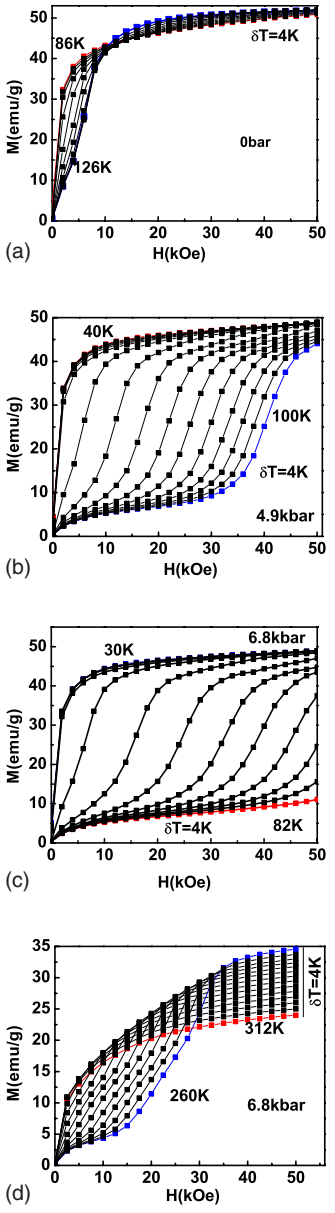

FIG. 3. (Color online) Magnetization isotherms of $SmMn_2Ge_2$ at temperatures near T_1 under a pressure of (a) 0 bar, (b) 4.9 kbar, (c) 6.8 kbar, and (d) M-H isotherms near T_2 in a pressure of 6.8 kbar.

low fields and a near saturation at high fields. The saturation magnetization (M_s) for all the pressures has been determined by plotting M vs $1/H$ in the high-field region and extrapolating the M values to the H axis. The M_s values of all the three pressures are found to be about $3.7\mu_B$/f.u. It should be noted that Tomka *et al.*[15] have reported that the Mn moment is about $3\mu_B$ and the Sm moment is in the range of 0.65 to $0.3\mu_B$ in $SmMn_2Ge_2$.

Figures 3(a)–3(c) show the M-H isotherms of $SmMn_2Ge_2$ obtained near the transition temperature T_1 in different pressures, whereas Fig. 3(d) shows the isotherms near T_2 obtained at the highest pressure. In plots shown in Figs. 3(a)–3(c), it can be seen that at the lowest temperatures in each case, the magnetization behavior is ferromagnetic. However, at elevated temperatures, metamagnetic transition

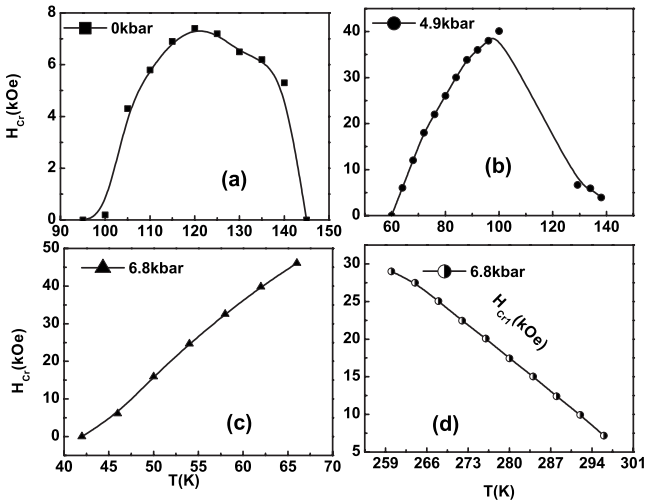

FIG. 4. Temperature dependences of the critical fields (H_{cr}) of metamagnetic transition in different pressures at temperatures close to T_1 and T_2.

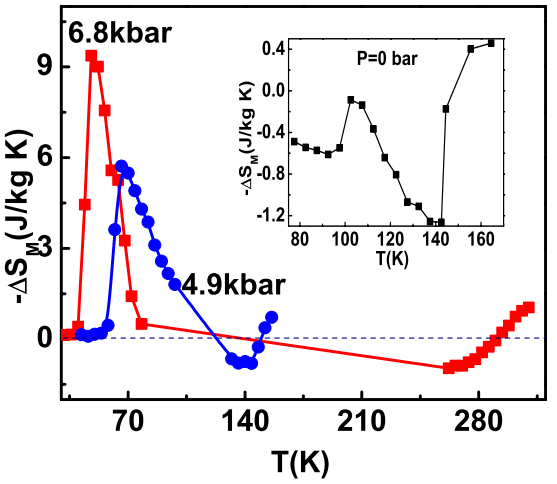

FIG. 5. (Color online) The temperature dependence of isothermal magnetic entropy change ($-\Delta S_M$) in $SmMn_2Ge_2$ at 50 kOe and in different pressures. Inset shows the entropy change at ambient pressure for the same field change.

from AFM to FM state is observed. It can be seen from Fig. 3(d) that the magnetic state changes from AFM to FM, as the temperature is increased from 260 to 312 K. It is also clear from Fig. 3(d) that, in low fields, the magnetization increases with the increase in temperature, whereas the trend gets reversed as the field exceeds the critical field needed for metamagnetic transition. These variations are consistent with the *M-T-P* data presented in Figs. 1 and 2.

Figure 4 shows the temperature variation of the critical field of $SmMn_2Ge_2$ in the temperature regions near T_1 and T_2. The transitions (at T_1 and T_2) in $SmMn_2Ge_2$ occur when the critical field H_{cr} is applied. From these figures, it is clear that critical fields for the transition at T_1 increase with increase in the pressure, which is due to the strengthening of the AFM order as a result of reduced Mn-Mn bond length. Applied pressure reinforces the interlayer Mn-Mn antiferromagnetic exchange interaction and leads to the increase of critical fields of metamagnetic transitions. The large increase in the critical fields with pressure can be noticed by comparing the Figs. 4(a)–4(c). The change in the sign of the slopes in Figs. 4(c) and 4(d) is due to the fact that the transition at T_1 causes a change from FM to AFM, while that at T_2 is from AFM to FM, as the temperature is increased.

The magnetocaloric effect has been calculated in terms of isothermal magnetic entropy change (ΔS_M) using the methods described elsewhere,[18,19] employing the magnetization isotherms obtained near the transition temperatures. Figure 5 shows the variation of magnetic entropy change in $SmMn_2Ge_2$ for a field change of 50 kOe under different pressures. The inset of this figure shows the MCE variation at ambient pressure for the same field change. As is clear from the inset, at ambient pressure, close to T_1, the entropy change is positive (negative MCE) with a magnitude, i.e., $(\Delta S_M)_{max}$, of about 0.6 J/kg K, whereas it is negative (positive MCE) close to T_2. Such a change in the sign of MCE has been reported by Koyama *et al.*[12] and Han *et al.*[13] as well. It was also observed that the entropy change (at ambient pressure) decreases with increase in field. At this point, it should be remembered that this compound shows a large positive magnetoresistance below 50 K.[8] Therefore, it is reasonable to assume that the origin of the MCE and MR behavior is the same in this compound.

Upon application of pressure, the sign of the entropy close to T_1 becomes negative, implying positive MCE. Furthermore, there is a significant increase in the magnitude of the entropy change. At a pressure of 4.9 kbar, $(\Delta S_M)_{max}$ is about 5.8 J/ kg K and it becomes 9.2 J/ kg K as the pressure is increased to 6.8 kbar. Therefore, it is clear that a pressure of 6.8 kbar has caused an increase in the entropy change by about 20 times, which must be due to the strengthening of the first-order nature of the magnetic transition at T_1 with pressure. Another point of interest is that, contrary to the ambient pressure data, the magnitude of the entropy change shows an increasing trend with increase in the applied field. Therefore, the nature of the MCE variation changes significantly on application of pressure. The entropy change associated with the transition at T_2 is also found to increase with pressure, though nominally. It should be noted that the sign of MCE at this temperature is positive, which is in agreement with the nature of the magnetic transition.

It is of interest to note that the competition of MCE of different signs in the ferromagnetic (below T_1) and antiferromagnetic (below T_2) phases gives rise to the anomalous pressure dependence. Such a competition is relieved by pressure as the transitions at T_1 and T_2 are driven in opposite directions. However, applied field has the opposite effect of bringing them together. This is interesting, as it is unlike most metamagnetic single-transition materials such as FeRh and Mn_3GaC.[22,23]

Figure 6 shows the temperature variation of magnetization in $GdMn_2Ge_2$ at 200 Oe under different applied pressures. It has been reported that at ambient pressure, this compound has a Neel temperature of 365 K, below which the Mn sublattice is antiferromagnetic and the Gd sublattice is not magnetically ordered. As the temperature is reduced to 95 K (T_C^{Gd}), the Gd sublattice gets ordered. Below this temperature

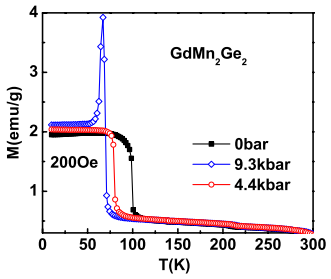

FIG. 6. (Color online) Temperature dependence of magnetization of $GdMn_2Ge_2$ in a magnetic field of 200 Oe for different pressures.

the coupling between Gd and Mn sublattices is AFM, since Gd is a heavy rare earth.[18] As in the case of $SmMn_2Ge_2$, the transition temperature (i.e., T_C^{Gd}) decreases with pressure with a rate of about 3.5 K/kbar, implying the strengthening of the AFM state. As can be seen, the transition seen in 9.3 kbar is different compared to that observed in lower pressures. While at lower pressures, the magnetic transition seems to enter a FM phase on cooling; at the highest pressure, it seems that the transition seems to be of AFM nature.

Figure 7 shows the temperature variation of the entropy change in $GdMn_2Ge_2$ for various applied pressures. As can be seen, the maximum value of the entropy change increases with pressure, but the rate of increase is much smaller compared to that in $SmMn_2Ge_2$. Another point to be noted is that the entropy change shows a minimum and the value corresponding to this minimum gradually decreases and becomes negative. The occurrence of the minimum is due to the increase in the entropy change at low temperatures. Wada *et al.*[24] have reported that, owing to the ferrimagnetic Gd-Mn coupling, the magnetic state of this compound is canted below T_C^{Gd}. This is because, with the application of field, the net field acting on the Mn moments decreases and therefore the moments tend to order antiferromagnetically. This results in a canted magnetic structure, causing considerable magnetic entropy change at temperatures below T_C^{Gd}. It is of importance to recall at this point that by virtue of the ferromagnetic

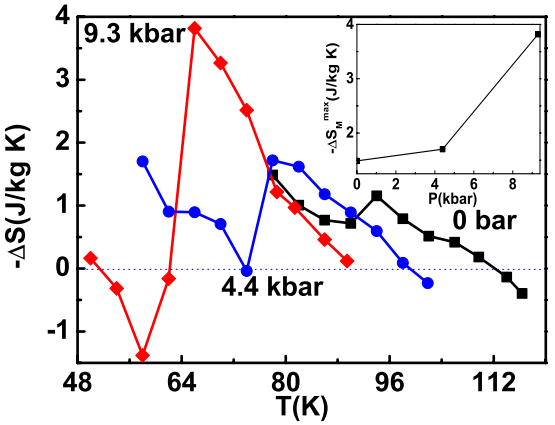

FIG. 7. (Color online) The temperature dependence of isothermal magnetic entropy change $(-\Delta S_M)$ in $GdMn_2Ge_2$ at 50 kOe and different pressures. Inset shows the variation of $(\Delta S_M)_{max}$ values with pressure.

coupling between Sm and Mn, such a behavior is absent in $SmMn_2Ge_2$. The dominance of the antiferromagnetic coupling of the Mn sublattice in $GdMn_2Ge_2$ explains the change in the sign of ΔS_M corresponding to the minimum at the highest pressure of 9.3 kbar. It is also evident from Fig. 7 that the temperature corresponding to the minimum (in ΔS_M) shifts to low temperatures, as the pressure is increased from the ambient value to 9.3 kbar, which is a reflection of the reduction in T_C^{Gd}.

IV. DISCUSSION

Tomka *et al.*[15] have reported the temperature-pressure phase diagram of $SmMn_2Ge_2$ using the ac magnetic susceptibility, neutron diffraction, and NMR studies under pressure. They have shown that the low-temperature (re-entrant) FM state is not the same for ambient pressure and elevated pressures. They suggest that the magnetic state of the re-entrant phase at ambient pressure consists of a net FM component $(\sim 2\mu_B)$ in the (001) planes along with an AFM component $(\sim 2.2\mu_B)$ in the c direction and a conical AFM coupling (interlayer) along $\langle 001 \rangle$. As the pressure is increased, the magnetic structure becomes commensurate with the nuclear structure, and as a result, the interlayer conical AFM changes to collinear AFM. Above T_1 (but $<T_2$), at ambient pressure, the magnetic state is such that it is conical AFM along $\langle 001 \rangle$, AFM component $(\sim 1.6\mu_B)$ within the (001) plane and FM component $(\sim 2\mu_B)$ along the c axis between the neighbors within the (001) plane. This structure is retained for pressures as high as about 9 kbar as well. In the high temperature FM phase, (i.e., above T_2), the magnetic structure is such that there is a FM component $(\sim 1.7\mu_B)$ along the c axis in the (001) plane, along with an AFM component $(\sim 1.9\mu_B)$ in the same plane as well as along the $<001>$ direction. Based on these results, they have suggested that RMn_2Ge_2 compounds with $d^a_{\text{Mn-Mn}} > 2.85$ Å have an AFM component in the (001) plane, whereas for $d^a_{\text{Mn-Mn}} < 2.85$ Å, the ordering in the (001) plane is purely ferromagnetic. They also have found that in the case of $SmMn_2Ge_2$ at ambient pressure among the three magnetically ordered phases, the AFM phase $(T_1 < T < T_2)$ has the smallest AFM component in the (001) planes and the largest net FM component in the c direction. Application of a field along (001) results in the conversion of the magnetic state from AFM to FM state by virtue of a metamagnetic transition, thereby increasing the AFM component in the (001) planes. Though the present study is on polycrystalline sample, this may be the reason for the unusual observation of a reduction in the magnitude of the positive MCE (negative entropy change) with increase in field in $SmMn_2Ge_2$ at ambient pressure. As mentioned earlier, close to T_1, this compound shows a large positive magnetoresistance. Therefore, the present MCE data as well as the reported magnetoresistance data seem to support the magnetic structure derived by Tomka *et al.* However, at higher pressures, the effect of applied field becomes less dominant and therefore, the MCE shows the normal trend of increase with increase in field. Application of pressure enhances the overall AFM in the Mn sublattice, which in turn strengthens the first-order nature of the transition at T_1. This explains

the large positive MCE close to T_1 seen at 6.8 kbar in $SmMn_2Ge_2$.

Since the rare-earth sublattice does not get affected considerably by the applied pressure, the changes in the magnetic and magnetocaloric properties of $GdMn_2Ge_2$ must also be dictated by the Mn sublattice, as in the case of $SmMn_2Ge_2$. The difference in the M-T behavior seen at 9.3 kbar in Fig. 6 may be attributed due to the lattice contraction at the highest pressure, which causes the $d^a_{\text{Mn-Mn}}$ distance to fall below the critical value, thereby causing a change in the magnetic coupling of the intralayer Mn-Mn moments from noncollinear FM to collinear FM. This reinforces the Gd-Mn ferrimagnetic coupling and hence the peak at the highest pressure. For the ambient pressure and 4.9 kbar, because of the noncollinear FM of the intralayer Mn moments, the low-temperature magnetization is dictated by the Gd moments and the magnetic ordering below T_C^{Gd} is predominantly ferromagnetic. The observation that the entropy change with pressure in $GdMn_2Ge_2$ is only nominal as compared to that in $SmMn_2Ge_2$ reflects the fact that the change in the interlayer as well as intralayer exchange coupling strengths is relatively less in $GdMn_2Ge_2$. This proposition is also confirmed by the lower values of dT_C^{Gd}/dP in this compound, as compared to the dT_1/dP values seen in $SmMn_2Ge_2$.

V. CONCLUSIONS

Considering the results obtained in $SmMn_2Ge_2$ and $GdMn_2Ge_2$, it can be seen that the effect of pressure is to change the interlayer and intralayer Mn-Mn exchange strengths, which leads to variation in the AFM and FM components in the Mn sublattice. The magnetic and magnetocaloric results are in agreement with the magnetic phase diagrams reported for these two compounds. The fact that Sm-Mn coupling is ferromagnetic and Gd-Mn coupling is ferrimagnetic (antiferromagnetic) plays a crucial role in determining their magnetic and magnetocaloric properties, both at ambient as well as under applied pressures.

*Corresponding author; suresh@phy.iitb.ac.in

[1] K. A. Gschneidner, Jr., V. K. Pecharsky, and A. O. Tsokol, Rep. Prog. Phys. **68**, 1479 (2005).
[2] A. M. Tishin and Y. I. Spichkin, *The Magnetocaloric Effect and Its Applications* (IOP, New York, 2003).
[3] V. K. Pecharsky and K. A. Gschneidner, Jr., Phys. Rev. Lett. **78**, 4494 (1997).
[4] S. Gama, A. A. Coelho, A. de Campos, A. M. G. Carvalho, F. C. G. Gandra, P. J. von Ranke, and N. A. de Oliveira, Phys. Rev. Lett. **93**, 237202 (2004).
[5] H. Wada and Y. Tanabe, Appl. Phys. Lett. **79**, 3302 (2001).
[6] H. Fujii, T. Okamoto, T. Shigeoka, and N. Iwata, Solid State Commun. **53**, 715 (1985).
[7] H. Fujii, M. Isoda, T. Okamoto, T. Shigeoka, and N. Iwata, J. Magn. Magn. Mater. **54-57**, 1345 (1986).
[8] E. V. Sampathkumaran, P. L. Paulose, and R. Mallik, Phys. Rev. B **54**, R3710 (1996).
[9] H. Wada, Y. Tanabe, K. Hagiwara, and M. Shiga, J. Magn. Magn. Mater. **218**, 203 (2000).
[10] T. Fujiwara, H. Fujii, and T. Shigeoka, Phys. Rev. B **63**, 174440 (2001).
[11] A. Barla, J. P. Sanchez, B. Malaman, B. P. Doyle, and R. Rüffer, Phys. Rev. B **69**, 220405(R) (2004).
[12] K. Koyama, S. Miura, H. Okada, T. Shigeoka, S. Fujieda, A. Fujita, K. Fukamichi, and K. Watanabe, J. Alloys Compd. **408-412**, 118 (2006).
[13] Z. Han, H. Wu, D. Wang, Z. Hua, C. Zhang, B. Gu, and Y. Du, J. Appl. Phys. **100**, 043908 (2006).
[14] S. Chaudhary, M. K. Chattopadhyay, K. J. Singh, S. B. Roy, P. Chaddah, and E. V. Sampathkumaran, Phys. Rev. B **66**, 014424 (2002).
[15] G. J. Tomka, C. Ritter, P. C. Riedi, Cz. Kapusta, and W. Kocemba, Phys. Rev. B **58**, 6330 (1998).
[16] E. G. Gerasimov, N. V. Mushnikov, and T. Goto, Phys. Rev. B **72**, 064446 (2005).
[17] K. Koyama, S. Miura, H. Okada, T. Shigeoka, and K. Watanabe, Mater. Trans. **48**, 451 (2007).
[18] Pramod Kumar, K. G. Suresh, A. K. Nigam, and S. K. Malik, J. Appl. Phys. **101**, 013908 (2007).
[19] Pramod Kumar, K. G. Suresh, A. K. Nigam, and S. K. Malik, J. Appl. Phys. **103**, 013909 (2008).
[20] T. Fujiwara, H. Fujii, and T. Shigeoka, Phys. Rev. B **63**, 174440 (2001).
[21] R. Welter, G. Venturini, E. Ressouche, and B. Malman, J. Alloys Compd. **218**, 204 (1995).
[22] M. P. Annaorazov, S. A. Nikitin, A. L. Tyurin, K. A. Asatryan, and A. Kh. Dovletov, J. Appl. Phys. **79**, 1689 (1996).
[23] M. H. Yu, L. H. Levis, and A. R. Moodenbaugh, J. Appl. Phys. **93**, 10128 (2003).
[24] H. Wada, H. Yoshioka, and T. Goto, J. Phys.: Condens. Matter **14**, L687 (2002).