Metamagnetic-transition-induced giant magnetoresistance in $Mn_2Sb_{1-x}Sn_x$ ($0<x\leq0.4$) compounds

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In $Mn_2Sb_{1-x}Sn_x$ ($0<x\leq0.4$) compounds, a metamagnetic transition from antiferromagnetic to ferrimagnetic can be induced by an external field, with which a giant magnetoresistance (GMR) effect is associated. The largest GMR ratio of 60% for $Mn_2Sb_{0.85}Sn_{0.15}$ is achieved at 142 K at a field of 5 T. The field dependence of GMR and the magnetization of $Mn_2Sb_{0.6}Sn_{0.4}$ confirm that the field-induced metamagnetic transition gives rise to the GMR effect in the system. The origin of the GMR effect is discussed in terms of the reconstruction of Fermi surface due to the collapse of the super zone gap after the metamagnetic transition.

Since the discovery of giant magnetoresistance (GMR) in antiferromagnetically coupled Fe/Cr multilayers,[1] the GMR effect has attracted much interest, which has been subsequently observed in many multilayer systems,[2–4] and granular films.[5,6] The GMR effect has also been found in rare-earth intermetallic compounds $NdCu_2$, $SmMn_2Ge_2$, $Ce(Fe_{1-x}Co_x)_2$, in $3d$ compound FeRh, and uranium compounds UNiGa, UPdIn, etc.[7] A great effort has been made in order to increase the GMR ratio which is usually low in the compounds, to increase the temperature of the antiferromagnetic ordering, and to lower the magnetic field for onset of the GMR.[7] It is also interesting to exploit the GMR in new intermetallic systems with interesting mechanisms of magnetotransport. In this work, we demonstrate the phase transitions and the GMR effects in a series of $3d$ compounds $Mn_2Sb_{1-x}Sn_x$ ($0<x\leq0.4$), and discuss in detail the origin of these phenomena.

All compounds of $Mn_2Sb_{1-x}Sn_x$ ($0<x\leq0.4$) were prepared by melting appropriate metals with purity higher than 99.9% in a magnetocontrolled arc furnace under a high purity argon atmosphere. An excess (10%) of Mn over the stoichiometric amount was added to compensate for the mass loss during melting. All the ingots were annealed at 700 °C for 62 h and gradually cooled to room temperature. X-ray diffraction studies at room temperature were carried out to certify that all the samples display peaks characteristic for Cu_2Sb-type structure with minor Mn_3Sn_2 as impurity phase (less than 5%). Magnetic properties were measured using a superconducting quantum interference device magnetometer in applied fields up to 5 T in the temperature range from 50 to 300 K. Magnetotransport properties were measured using a standard four-probe dc method, while warming.

It has been reported that Mn_2Sb is a ferrimagnet. In the crystal structure there are two crystallographically different Mn atoms, i.e., Mn1 and Mn2. According to neutron diffraction measurements the magnetic structure can be regarded as a stacking of triple layers of Mn2-Mn1-Mn2 with antiparallel magnetic moments on Mn1 and Mn2 sites in both the ferrimagnetic (FI) and antiferromagnetic (AF) states.[8] The moments of all triple layers are parallel in the FI state, whereas the arrangement is antiparallel in the AF state.

The substitution of a number of elements (Co, Cr, Zn, and Cu) for Mn, as well as of As and Ge for Sb results in the appearance of a first-order phase transition from the FI to the AF structure as the temperature decreases.[9] Figure 1 shows the temperature dependence of the resistivities $\rho(T)$ of $Mn_2Sb_{1-x}Sn_x$ ($0\leq x\leq0.4$) measured in a zero field and an applied field of 5 T. The resistivity of Mn_2Sb decreases monotonically with decreasing temperature, while the resistivities of other samples first decrease, then rise abruptly at a certain temperature reaching a maximum, and finally decrease with decreasing temperature. The present results for the resistivities in the zero field are in good agreement with the previous work.[10] The considerable temperature hysteresis of the electrical resistance was reported.[10] It is evident that a
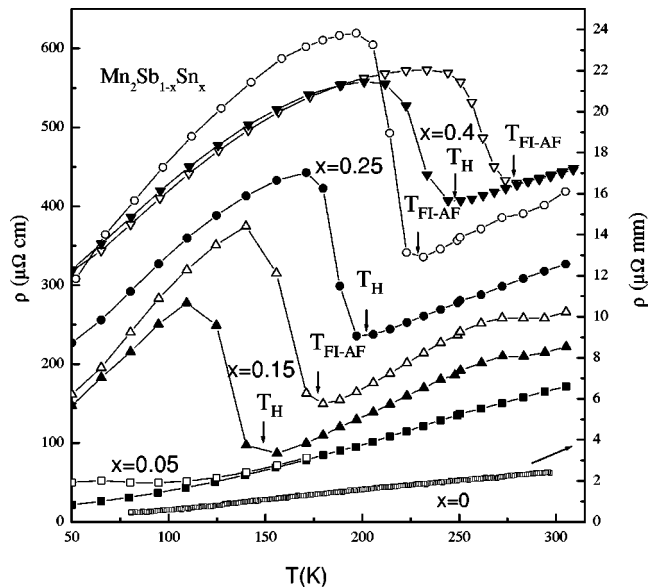

FIG. 1. Temperature dependence of the resistivities of $Mn_2Sb_{1-x}Sn_x$ compounds at zero field (the open symbols) and at a magnetic field of 5 T (the solid symbols).
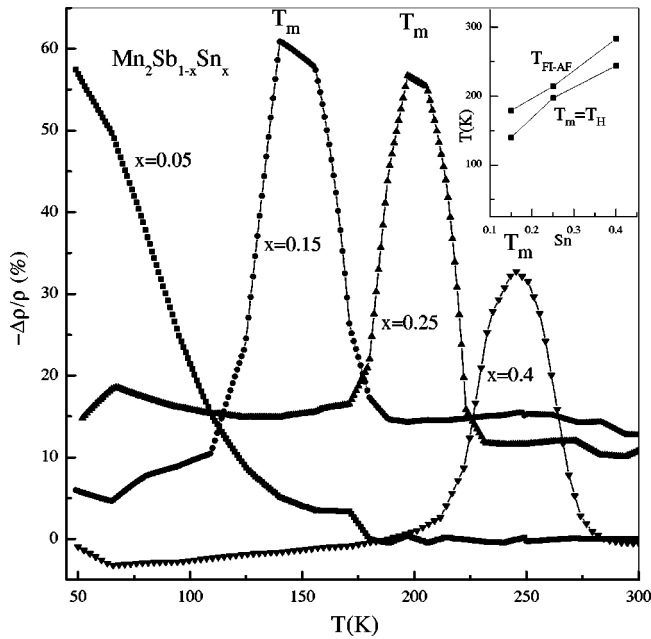
FIG. 2. Temperature dependence of the magnetoresistance of $Mn_2Sb_{1-x}Sn_x$ compounds at a magnetic field of 5 T.

partial substitution of Sb by Sn causes the transition from the FI to the AF phase at a certain temperature (T_{FI-AF}), which is in good agreement with earlier work,[10,11] where the magnetic transition was verified by using direct magnetic measurements and the Mössbauer effect. The transition temperature is quite sensitive, not only to the applied magnetic field, but also to the Sn concentration. The FI-AF transition temperature increases with Sn concentration from 94 K for $x=0.05$ to 283 K for $x=0.4$ in the zero field. The magnetic field shifts the FI-AF transition temperature T_{FI-AF} towards lower temperatures correspondingly.

The AF phase collapses under an applied magnetic field in a certain temperature range. Namely, a magnetic-field-induced metamagnetic transition takes place in $Mn_2Sb_{1-x}Sn_x$ ($0 < x \le 0.4$). As a result, a negative MR is observed in these compounds. Figure 2 shows the temperature dependence of the GMR ratio $\Delta\rho/\rho$ [$=(\rho_H-\rho_0)/\rho_0$] of $Mn_2Sb_{1-x}Sn_x$ ($0 < x \le 0.4$) compounds at a field of 5 T. The magnetoresistance $\Delta\rho/\rho$ is small at low temperatures, since the compound is in the stable AF state at 5 T. When temperature increases up to a certain temperature, the AF structure becomes unstable in the field of 5 T. Namely, a metamagnetic transition occurs, resulting in a rapid increase of $-\Delta\rho/\rho$ of all the compounds. This occurs until a peak temperature T_m, which is about 140, 197, and 244 K for $x=0.15$, 0.25, and 0.4, respectively, just the same as the phase transition temperature T_H (see Fig. 1) at the field of 5 T. Above T_m, $\Delta\rho/\rho$ drops rapidly with increasing temperature. It is seen from the inset of Fig. 2 that the value of T_m (same as T_H) for each compound is slightly lower than its T_{FI-AF}, due to the effect of the magnetic field. The largest values of $\Delta\rho/\rho$ are -57%, -60%, -56%, and -36%, respectively, for $x=0.05$, 0.15, 0.25, and 0.4. To our knowledge, it is the first report on the GMR effect in $Mn_2Sb_{1-x}Sn_x$. Moreover, the

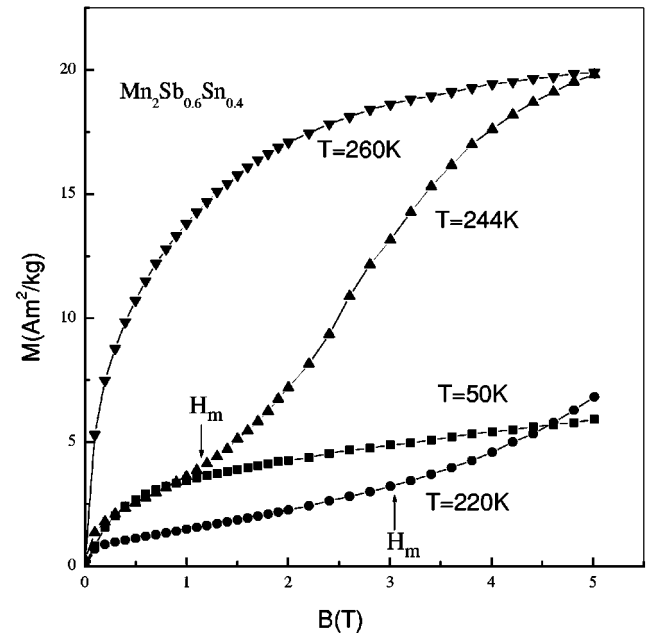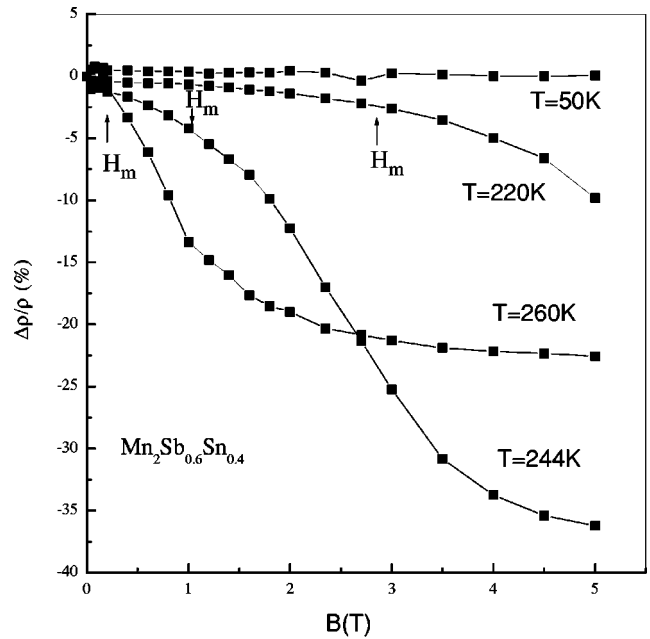FIG. 3. Field dependence of (a) the magnetoresistance and (b) the magnetization of a $Mn_2Sb_{0.6}Sn_{0.4}$ compound at different temperatures.

large MR changes in many antiferromagnetic ordered compounds reported previously have been confined to very low temperatures because of their low values of Néel temperature T_N.[7] As seen from Fig. 3, the transition from AF to FI can induced by moderate fields (about 1 T) at 244 K. Therefore, $Mn_2Sb_{0.6}Sn_{0.4}$ might be more attractive for applications, compared to other compounds.

To better understand the magnetic transport behavior, the magnetic field dependence of the GMR ratio $\Delta\rho/\rho$ and the magnetization of $Mn_2Sb_{0.6}Sn_{0.4}$ at different temperatures are represented in Figs. 3(a) and 3(b). Between 50 and 244 K, the $\Delta\rho/\rho$ is negligibly small at low fields, then it develops
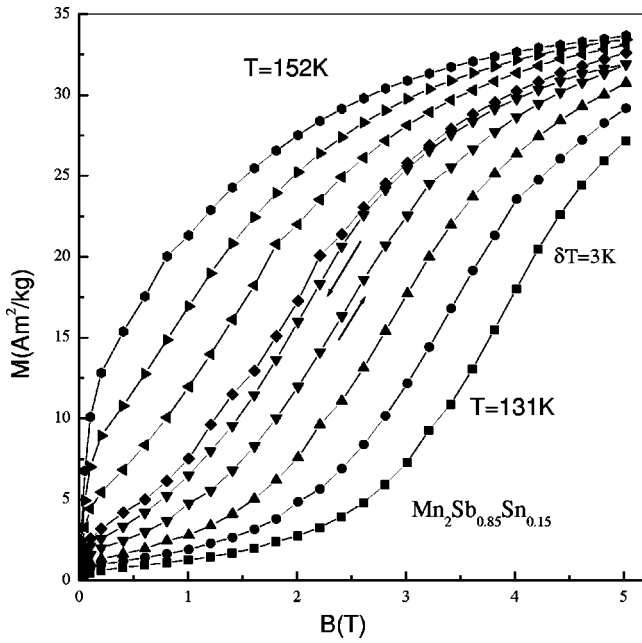
FIG. 4. Field dependence of the magnetization of a $Mn_2Sb_{0.85}Sn_{0.15}$ compound at different temperatures.

drastically around a critical field H_m, showing a negative contribution. The small $\Delta\rho/\rho$ is indicative of a stable antiferromagnetic (AF) state and the development of a significant negative GMR is associated with the metamagnetic transition to the FI state as mentioned above. A similar transition related to GMR was earlier observed in $Ce(Fe,Al)_2$, $Ce(Fe,Ru)_2$, UCu_2Ge_2, and Mn_3CaC compounds.[12–15] At 260 K, $\Delta\rho/\rho$ shows a tendency to saturate at a field of 3 T. In the present measurement, the largest value for $\Delta\rho/\rho$ at 244 K of $Mn_2Sb_{0.6}Sn_{0.4}$ is 36% in a magnetic field of 5 T, although it does not saturate yet. Moreover, from Fig. 3(a), the critical field H_m for the metamagnetic transition becomes smaller with increasing temperature.

The magnetic field dependence of magnetization shown in Fig. 3(b) for $Mn_2Sb_{0.6}Sn_{0.4}$ is in good agreement with the field dependence of magnetoresistance above. Between 50 and 244 K, the magnetization rises initially and then increases gradually after a certain field. Further applying a higher field (depending on temperature), the magnetization starts increasing rather rapidly, showing a metamagnetic transition. The critical field at which the metamagnetic transition starts coincides with H_m. No saturation in magnetization is observed up to fields of 5 T in this temperature range (the maximum value of magnetization approaches 20 Am^2/kg). The same measurement for $Mn_2Sb_{0.85}Sn_{0.15}$ compound was performed to further confirm the metamagnetic transition (see Fig. 4). It is clearly seen that the metamagnetic transition field decreases with increasing temperature, and a hysteresis was observed for all the magnetization curves (only an example of the hysteresis is shown in Fig. 4) as expected for a first order transition. Moreover, the saturation magnetization of $Mn_2Sb_{0.85}Sn_{0.15}$ is larger than that of $Mn_2Sb_{0.6}Sn_{0.4}$, which is in agreement with early experiment[10] indicating that the substitution of Sb atoms by

Sn atoms in the nonmagnetic sublattice leads to the quantitative redistribution of the sublattice magnetizations of Mn1 and Mn2. With the Sn concentration increasing, the antiferromagnetic exchange interaction of Mn1 and Mn2 becomes stronger, to make the phase transition temperature shift to high temperature (see Fig. 1).

The sharp rise of resistivities below AF ordering temperature in $Mn_2Sb_{1-x}Sn_x$ compounds is tentatively ascribed to super-zone gap as a consequence of an additional periodicity in the AF state.[16–18] If this is the case, the gap should be suppressed when the AF order is destroyed by a sufficiently high magnetic field. It is seen from above analysis that the field for rapid decrease/increase of the resistivity/GMR is in good agreement with the metamagnetic transition field in the $M(B)$ curve of the $Mn_2Sb_{0.6}Sn_{0.4}$ compound. Therefore the superzone gap is eliminated by the metamagnetic transition from the AF state to a field-induce FI state, similar to the effects observed in $CeSbNi_x$,[16] $Ce(Fe_{1-x}Co_x)_2$,[17] $Eu_{14}MnBi_{11}$,[18] $Ce(Fe_{0.98}Ru_{0.07})_2$,[13] and $Mn_{1.9}Cr_{0.1}Sb$.[19] The neutron diffraction data showed that the period of the AF structure along the tetragonal axis for $Mn_{1.9}Cr_{0.1}Sb$ is equal to a double period of the FI structure for Mn_2Sb along the same axis.[20] It was reported[21] that the abrupt increase of the electrical resistivity below the AF ordering temperature is due to the large decrease of the density of states (DOS) near the Feimi level, in accordance with the band structure calculations of Mn_2Sb and the specific heat of $Mn_{1.9}Cr_{0.1}Sb$,[22] $Mn_{1.82}Co_{0.18}Sb$,[21] and Mn_2Sb.[21] The $3d$ electrons of Mn in Mn_2Sb have an itinerant character, and the Fermi level lies between two-peak structures on the DOS curve.[8,21] Hence, the change in the d-electron concentration due to substitution should lead to a significant change in the DOS, resulting in the change of the Magnetic order. Similar to FeRu,[23] $Ce(Fe_{1-x}Co_x)_2$,[24] $UNiGa$,[25,26] and Mn_3GaC (Ref. 27) compounds, the electronic origin of the AF-FI transition of Mn_2Sb-based compounds is that the DOS near the Fermi level is changed during the phase transition.

It is known that the magnetic structure of $Mn_2Sb_{1-x}Sn_x$ compounds is similar to that of magnetic multilayer systems.[8] One possible mechanism for the origin of GMR in $Mn_2Sb_{1-x}Sn_x$ compounds is a spin-dependent scattering mechanism leading to a reduction of the relaxation time in the AF state, as commonly considered in the magnetic multilayer systems. As mentioned above, another possibility is the variation of the Fermi surface when crossing the metamagnetic transition (for example, due to the disappearance of the superzone gap resulting from the FI ordering), which yields the variation of the DOS near the Fermi surface. According to the mechanism of the FI-AF transition[28] and the abrupt increase of resistivities below the AF ordering temperature, the latter mechanism should be dominant in $Mn_2Sb_{1-x}Sn_x$ compounds.

In summary, we report on a system of intermetallic compounds $Mn_2Sb_{1-x}Sn_x$ ($x=0.05$, 0.15, 0.25, and 0.4) with large GMR effects. The largest MR ratio is 60% for $Mn_2Sb_{0.85}Sn_{0.15}$ at 142 K in a magnetic field of 5 T. Moreover, the $Mn_2Sb_{0.6}Sn_{0.4}$ compound has a high antiferromagnetic ordering temperature of 283 K, a low metamagnetic transition field of 1 T at 244 K, and a comparatively large

GMR of 25% at a field of 3 T at 244 K, which indicate that it may have potential application as a material for GMR devices. Field-induced metamagnetic transition gives rise to GMR in the system, and the origin of the GMR effect is discussed in terms of the reconstruction of Fermi surface due to the collapse of the superzone gap after the metamagnetic transition.

[1] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Frederich, and J. Chazeles, Phys. Rev. Lett. **61**, 2472 (1988).

[2] S. S. P. Parkin, Z. G. Li, and D. J. Smith, Appl. Phys. Lett. **58**, 2710 (1991).

[3] S. S. P. Parkin, R. Bhadra, and K. P. Roche, Phys. Rev. Lett. **66**, 2152 (1991).

[4] F. Petroff, A. Barthelemy, D. H. Mosca, D. K. Lottis, A. Fert, P. A. Schroeder, W. P. Pratt, R. Loloee, S. Lequien, Phys. Rev. B **44**, 5355 (1991).

[5] J. Q. Xiao, J. S. Jiang, and C. L. Chien, Phys. Rev. Lett. **68**, 3749 (1992).

[6] A. E. Berkowitz, M. J. Carey, J. R. Mitchell, A. P. Young, S. Zhang, F. E. Spada, F. T. Parker, A. Hutten, and G. Thomas, Phys. Rev. Lett. **68**, 3745 (1992).

[7] V. Sechovsky, L. Havela, K. Prokes, H. Nakotte, F. R. de Boer, and E. Brück, J. Appl. Phys. **76**, 6913 (1994).

[8] J. H. Wijngaard, C. Haas, and R. A. de Groot, Phys. Rev. B **45**, 5395 (1992).

[9] T. A. Bither, P. H. L. Walter, W. H. Cloud, T. J. Swoboda, and P. E. Bierstedt, J. Appl. Phys. **33**, 346 (1962).

[10] V. M. Ryzhkovsk, V. P. Dymont, and Z. L. Erofeenko, Phys. Status Solidi A **130**, 163 (1992).

[11] C. Blaauw, G. R. Hackay, and W. Leiper, J. Magn. Magn. Mater. **7**, 234 (1978).

[12] S. Radha, S. B. Roy, A. K. Nigam, and G. Chandra, Phys. Rev. B **50**, 6866 (1994).

[13] H. P. Kunkel, X. Z. Zhou, P. A. Stampe, J. A. Cowen, and G. Williams, Phys. Rev. B **53**, 15 099 (1996).

[14] A. K. Nigam, S. B. Roy, and G. Chandra, Phys. Rev. B **49**, 1127 (1994).

[15] S. Y. Zhang, P. Zhao, Z. H. Cheng, R. W. Li, J. R. Sun, H. W. Zhang, and B. G. Shen, Phys. Rev. B **64**, 212404 (2000).

[16] M. H. Jung, D. T. Adroja, N. Kikugawa, T. Takabatake, and I. Oguro, Phys. Rev. B **62**, 13 860 (2000).

[17] H. Fukuda, H. Fujii, and H. Kamura, Phys. Rev. B **63**, 054405 (2001).

[18] J. Y. Chan and S. M. Kauzlarich, Phys. Rev. B **57**, R8103 (1998).

[19] Y. Nagata, T. Hagii, S. Yashiro, H. Samata, and S. Abe, J. Alloys Compd. **292**, 11 (1999).

[20] T. J. Swoboda, W. H. Cloud, T. A. Bither, M. S. Sadler, and H. S. Jarret, Phys. Rev. Lett. **4**, 509 (1960).

[21] M. I. Bartashevich, T. Goto, T. Tomita, N. V. Baranov, S. V. Zemlyanski, G. Hilscher, and H. Michor, Physica B **318**, 198 (2002).

[22] E. Gratz, G. Hilscher, and R. Resel, J. Alloys Compd. **187**, L7 (1992).

[23] M. P. Annaorazov, S. A. Nikitin, A. L. Tyurin, K. A. Asatryan, and A. Kh. Dovletov, J. Appl. Phys. **79**, 1689 (1996).

[24] H. Wada, T. Harada, and M. Shiga, J. Phys.: Condens. Matter **9**, 9347 (1997).

[25] Y. Kobayashi, Y. Aoki, H. Sugawara, and H. Sato, Phys. Rev. B **54**, 15 330 (1996).

[26] L. Havela, V. Sechovsky, Y. Aoki, Y. Kobayashi, H. Sato, K. Prokes, M. Mihalik, and A. A. Menovsky, J. Appl. Phys. **81**, 5778 (1997).

[27] K. Kamishima, T. Goto, H. Nakagawa, N. Miura, M. Ohashi, N. Mori, T. Sasaki, and T. Kanomata, Phys. Rev. B **63**, 024426 (2001).

[28] E. P. Wohlfarth, Phys. Lett. **4**, 83 (1963).