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LETTER TO THE EDITOR

Novel magnetic phase transitions and magnetoresistance of $GdMn_2Ge_2$

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Abstract

The magnetization and magnetoresistance (MR) were measured for $GdMn_2Ge_2$, which shows a first-order transition from a low-temperature ferrimagnetic (Fi) state to a high-temperature antiferromagnetic (AF) state at $T_1 = 95$ K. The field-induced transition from the Fi state to the high-field state was observed below T_1. The transition field decreases with increasing temperature and it falls to zero at T_1 for $H \perp c$, while the transition disappears above 90 K for $H \parallel c$. A large positive MR jump was observed associated with the field-induced transition for $H \perp c$. These results suggest that this may be the transition from the Fi to AF state caused by a magnetic field.

Ternary RMn_2Ge_2 compounds, where R is a rare-earth element, crystallize in the tetragonal $ThCr_2Si_2$-type structure, which is characterized by stacked R–Ge–Mn–Ge–R layers along the c-axis. In these compounds, R and Mn atoms possess magnetic moments and the compounds exhibit relatively high magnetic ordering temperatures [1]. The intralayer Mn–Mn interaction is the strongest, giving a ferromagnetic (F) arrangement of Mn moments in the c-plane. The interlayer Mn–Mn interaction strongly depends on the Mn–Mn distance in the c-plane, R_{Mn-Mn}. Early studies have revealed that the Mn sublattice in RMn_2Ge_2 with R_{Mn-Mn} larger than the critical value, $R_C = 0.285$ nm, shows the F ordering, whereas that with $R_{Mn-Mn} < R_C$ shows the antiferromagnetic (AF) ordering [1, 2]. In the case of R = Gd, Tb and Dy, the interlayer Mn–Mn interaction is negative and R_{Mn-Mn} is close to R_C. Magnetization measurements of $GdMn_2Ge_2$ have shown that the Mn moments are ordered antiferromagnetically below $T_N = 365$ K, where the Gd moments remain disordered, and a first-order transition from an AF state to a ferrimagnetic (Fi) state takes place at $T_1 = 95$ K with decreasing temperature [3]. In the Fi state, the Gd moments are ordered ferromagnetically along the c-axis and all the Mn moments are aligned antiparallel to the Gd moments. These experimental results have

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been explained by taking account of the competition between the Gd–Mn interaction and the Mn–Mn interaction [4]. On lowering the temperature, the negative R–Mn interaction develops and it overcomes the energy difference between F and AF states of the Mn sublattice, which leads to the Fi state.

Another important feature of $GdMn_2Ge_2$ is a field-induced magnetic transition below T_1. Kobayashi *et al* [5] found that the Fi state is replaced by a new magnetic phase in high magnetic fields, when a magnetic field is applied perpendicular to the c-axis at 77 K. Sokolov *et al* [6] have also observed this field-induced transition for aligned powder samples of $GdMn_2Ge_2$ at 4.2 K. The magnetic moment above the transition was estimated to be about 6 μ_B/fu, suggesting a canted spin structure in the high-field (HF) state. A similar field-induced transition from a Fi state to a HF state was reported for $DyMn_2Ge_2$ [7]. However, the magnetic structure of the HF state has not been manifested and the origin of the field-induced transition is still unclear.

In this letter, we report on the magnetization and magnetoresistance (MR) of $GdMn_2Ge_2$ single crystals. We observed a large positive MR associated with the field-induced transition below T_1. The results suggest that the HF state is identical to the AF state in a magnetic field.

Single crystals of $GdMn_2Ge_2$ were grown from In fluxes [8]. The pure constituents, of which the atomic ratio is R:Mn:Ge:In = 1:1:1:20, were placed in an alumina crucible and sealed under vacuum. This mixture was heated at 1100 °C and cooled to 500 °C at a rate of 7.5 °C h^{-1}. Then, the In fluxes were removed by a centrifuge at 500 °C. By this method, we were able to obtain thin plate-like single crystals, whose c-axis is perpendicular to the plate. The HF magnetization was measured in a pulsed magnetic field by an induction method with well-balanced pickup coils. The magnetization curves up to 5 T were measured using a commercial superconducting quantum interference device (SQUID). The MR measurements were performed by a four-probe method with an AC resistance bridge up to 8 T.

Figure 1 shows the magnetization (M) curves of $GdMn_2Ge_2$ at 4.2 K with a field applied parallel and perpendicular to the c-axis. The field-induced transition takes place for both field directions. For $H \parallel c$, the magnetization is saturated at a low field with a saturation magnetic moment of 3 μ_B/fu, which is in agreement with the previous reports [3, 5]. This fact indicates that the c-axis is the magnetization easy axis in the Fi state. The Mn moment is estimated to be 2 μ_B. For $H \perp c$, the transition field, H_C, is lower than that for $H \parallel c$. When we plot the M as a function of a reduced magnetic field, H/H_C, we have a universal magnetization curve for both field directions in a field range of $0.8 \leqslant H/H_C \leqslant 1.2$. These results indicate that the field-induced transition for $H \perp c$ is essentially the same as that for $H \parallel c$, i.e., the transition from the Fi state to the HF state along the field direction. The M-value just above H_C is about 5 μ_B/fu irrespective of the field direction, which is smaller than the theoretical value of the Gd^{3+} moment, 7 μ_B. Furthermore, the magnetization increases with increasing field with a large slope above H_C. These results support a canted spin structure in the HF state proposed previously [5, 6]. A step is recognizable in the field-induced transition in the M–H curve for $H \perp c$. Similar steps in the magnetization curves were recently reported by Fujiwara *et al* for $Gd_{0.925}La_{0.075}Mn_2Ge_2$ [9]. They claimed that the multisteps in the M–H curve are intrinsic for RMn_2Ge_2. In our case, however, the presence of the step strongly depends on the sample. At present, the origin of this magnetization step is an open question. We have studied the field-induced transition at finite temperatures. The temperature dependence of H_C is displayed in the inset of figure 1. For $H \perp c$, the transition was visible up to T_1. H_C decreases with increasing temperature and falls to zero at T_1. On the other hand, the field-induced transition is smeared for $H \parallel c$ above 90 K and vanishes at T_1 without H_C approaching zero.

The isothermal magnetization curves for $H \perp c$ near T_1 are depicted in figure 2. The magnetization curves below 93 K show a small remanent magnetization probably due to a slight deviation of the field direction from the c-axis, while no remanence was observed at

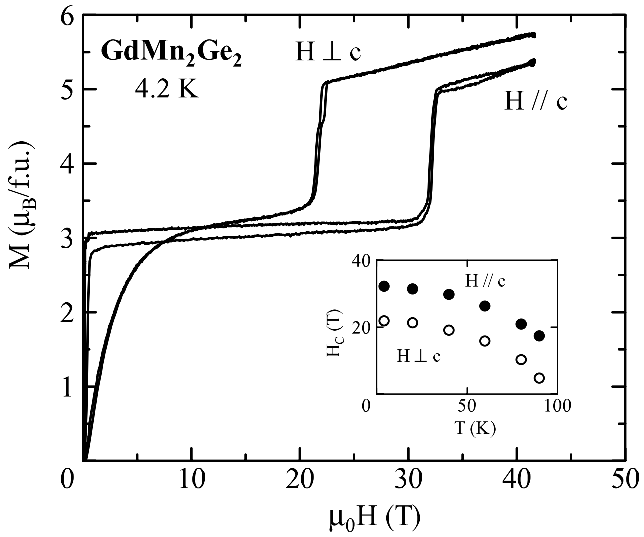

Figure 1. Magnetization curves of $GdMn_2Ge_2$ single crystals at 4.2 K with a field applied parallel and perpendicular to the c-axis. The inset shows the temperature dependence of the transition field, H_C, for $H \perp c$ and $H \parallel c$.

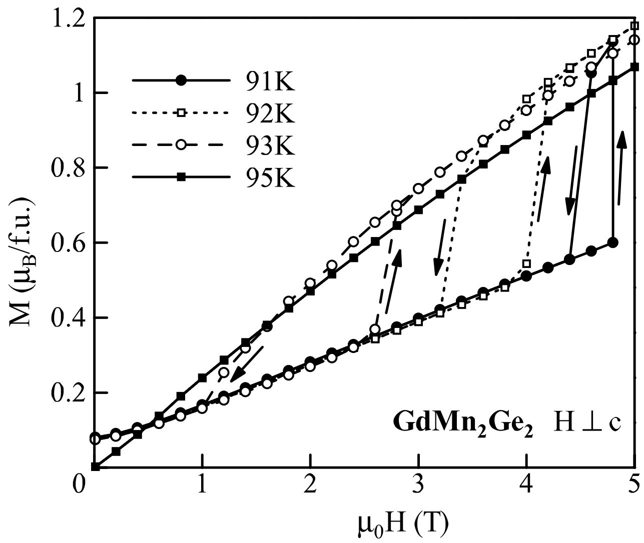

Figure 2. The M–H curves of $GdMn_2Ge_2$ for $H \perp c$ just below T_1.

95 K. Between 91 and 93 K, the magnetization curves show a jump at appropriate fields, indicating a transition to the HF phase. The transition was accompanied by a hysteresis and H_C goes to zero as the temperature is increased to T_1. Above H_C, M increases with increasing field like in the AF state. These results suggest that the HF state is identical to the AF state in a magnetic field.

To elucidate this point, we have measured the MR of $GdMn_2Ge_2$. The field dependence of the electrical resistivity, ρ, at various temperatures is shown in figure 3 with a field applied perpendicular to the c-axis. We found an abrupt increase of the resistivity in a magnetic

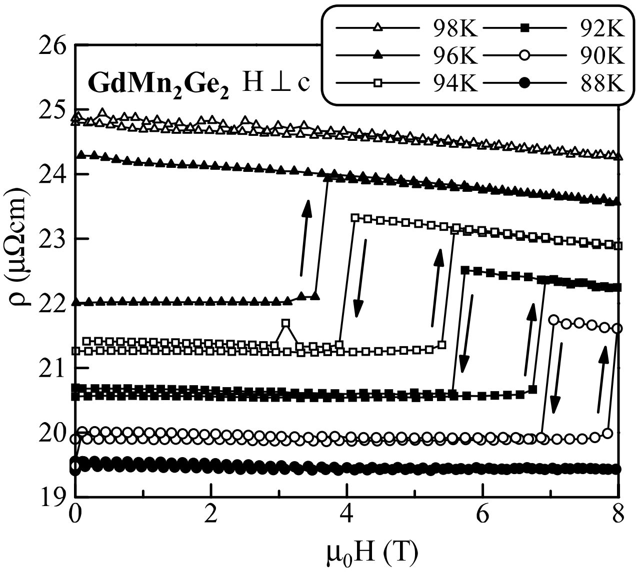

Figure 3. The MR of $GdMn_2Ge_2$ for $H \perp c$ just below T_1.

field in the temperature range of 90–96 K. This anomaly is clearly associated with the field-induced transition. A large hysteresis is consistent with the magnetization curves shown in figure 2. The relative change of resistivity at the transition field, $\Delta\rho/\rho(H_C)$, is about +10%. The prominent feature is a positive resistivity jump. In general, the MR jump due to a field-induced transition of magnetic compounds is negative. Typical examples are FeRh [10] and $ErCo_2$ [11]. The former compound undergoes a temperature-induced transition between AF and F states. Baranov *et al* [10] reported that $\Delta\rho/\rho(H_C)$ for $Fe_{49}(Rh_{0.9225}Pd_{0.06}Ir_{0.0175})_{51}$ is -60% at around 300 K. This giant negative MR is ascribed to the disappearance of a superzone gap at the Fermi level. The latter system exhibits a first-order Fi-to-paramagnetic (P) transition at $T_C = 32$ K. Just above T_C, $ErCo_2$ shows itinerant electron metamagnetism. In this case, the metamagnetic transition suppresses spin fluctuations, resulting in a considerable negative MR of $\Delta\rho/\rho(H_C) = -50\%$ [11]. Thus, the positive MR jump of the present system is very exceptional. Some rare-earth compounds are known to show multiple metamagnetic transitions. In such systems, a positive MR jump is sometimes observed during the transitions [12]. However, the MR finally decreases when the metamagnetic transitions are completed. This is similar to the spin-valve effect of multilayers [13]. Recently, Levin *et al* [14] reported a positive MR jump in $Gd_5Si_{1.5}Ge_{2.5}$, just above T_C. This compound undergoes a first-order magnetic transition at $T_C = 206$ K accompanied by a structural transition. The positive MR of the compound is attributable to the structural transition. On the other hand, the positive MR jump of $GdMn_2Ge_2$ is purely of magnetic origin. As described above, the HF state can be regarded as the AF state in a magnetic field. Thus, the field-induced transition switches the magnetic coupling between the Mn layers from the F type to the AF type. Such a change of magnetic structure causes a superzone gap at the Fermi level, leading to a reduction of the number of conduction electrons. We believe that this is the origin of a dramatic increase of the resistivity at H_C.

Higher resistance in the AF state than in the Fi state can be confirmed by the MR measurements just above T_1. When the magnetic field is applied parallel to the c-axis above

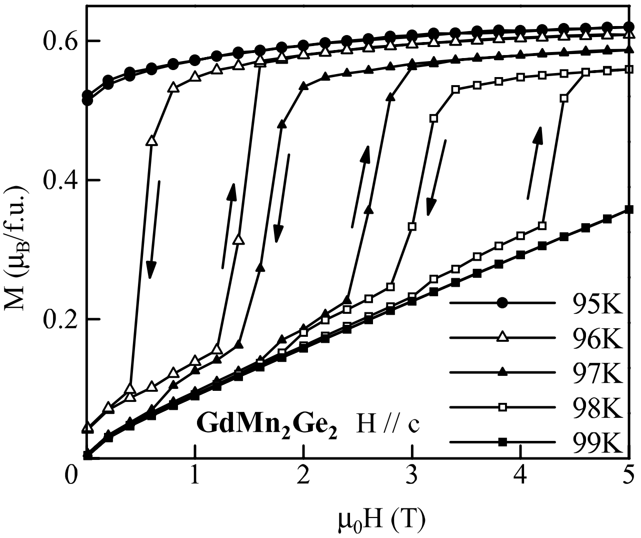

Figure 4. The M–H curves of $GdMn_2Ge_2$ for $H \parallel c$ above T_1.

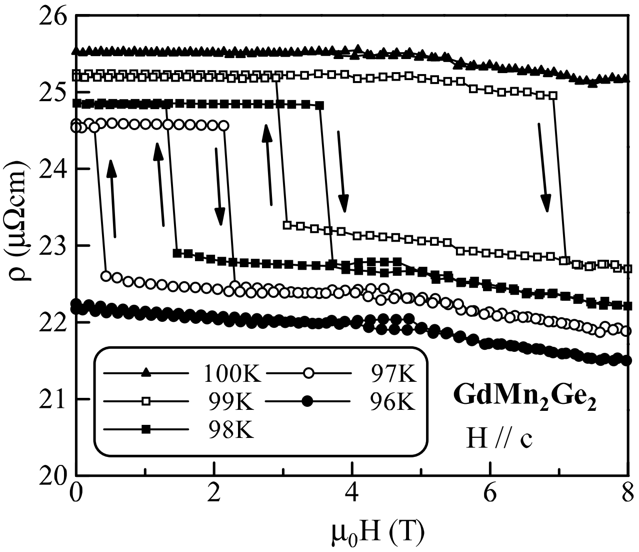

Figure 5. The MR of $GdMn_2Ge_2$ for $H \parallel c$ above T_1.

T_1, the compound exhibits a metamagnetic transition from the AF state to the Fi state, as shown in figure 4. The transition field increases as the temperature is increased, in contrast to the field-induced transition below T_1. The corresponding MR is displayed in figure 5 as a function of magnetic field. We observed a large negative MR jump associated with the metamagnetic transition. The absolute value of $\Delta\rho/\rho(H_C)$ is comparable to that observed in figure 3. These results indicate that the metamagnetic transition is the reverse of the field-induced transition process observed below T_1.

The present results suggest that the HF state is identical to the AF state in a magnetic field at $T \sim 90$ K. In other words, the transition from the Fi state to the AF state may be induced by a magnetic field in $GdMn_2Ge_2$ for $H \perp c$ just below T_1. If this is the case, this is a new type of magnetic transition. The field-induced transition can be explained by the competition of the R–Mn and Mn–Mn interactions. As described previously, R_{Mn-Mn} for $GdMn_2Ge_2$ is just below R_C and the AF structure of the Mn sublattice is barely realized below $T_N = 365$ K. In the Fi state, the negative Gd–Mn interaction aligns the Mn moments antiparallel to the Gd moments. When a magnetic field is applied to the Fi state, the direction of an applied field is opposite to the Mn moments. This indicates that the total field (=a molecular field + an applied field) acting on the Mn moments is decreased with increasing magnetic field, which destabilizes the Fi structure, leading to the AF coupling between the Mn layers. Near T_1, the Gd moments are thermally fluctuated and their contribution to the magnetization is small. This is why the HF state is essentially the same as the AF state in a magnetic field at high temperatures. At low temperatures, the Gd moments play a dominant role in the magnetization. Therefore, the Mn sublattice feels the substantial molecular field from the Gd moments in the HF state and, as a result, a canted spin structure is realized. The observed magnetic moment in the HF state is about 5 μ_B/fu just above H_C at 4.2 K. Assuming the Mn moment of 2 μ_B, we conclude that the Mn moment makes an angle of 30° with the negative field direction.

The M–H curves in figure 1 show that H_C for $H \parallel c$ is larger than that for $H \perp c$. This is ascribed to the magnetocrystalline anisotropy of the Mn sublattice. The magnetization easy axis is the c-axis in the present system. Since the single-ion anisotropy of the Gd ion is small, this magnetocrystalline anisotropy mainly comes from the Mn atoms. The field-induced transition is accompanied by an abrupt change of direction of the Mn moments. For $H \perp c$, the Mn moments lie in the c-plane below H_C, while they rise towards the c-axis in the HF state. In contrast, the Mn moments fall down towards the c-plane for $H \parallel c$. Clearly the magnetocrystalline anisotropy is favourable to the transition for $H \perp c$ and it reduces the critical field.

The field-induced transition was observed up to T_1 for $H \perp c$ but not for $H \parallel c$. We point out that the slope of the M–H curve in the AF state is larger than that in the Fi state for $H \perp c$ (see figure 2). When the magnetic field is applied perpendicular to the c-axis, the magnetic moments have to rotate towards the c-plane. However, the magnetocrystalline anisotropy prevents the Mn moments from rotating freely from the c-axis. Consequently, the Gd moments coupled antiparallel to the Mn moments give a small magnetization in the Fi state. On the other hand, the Gd moments are disordered in the AF state. Thus, the magnetization induced by a magnetic field in the AF state is larger than that in the Fi state for $H \perp c$.

In conclusion, $GdMn_2Ge_2$ exhibits the field-induced magnetic transition below T_1. Near T_1, the transition was observed only for $H \perp c$. A positive MR jump was detected associated with the transition. These results are indicative that this is the transition from the Fi state to the AF state induced by a magnetic field. Direct measurements of a magnetic structure, such as neutron diffraction measurements, are strongly desired to clarify the nature of this field-induced magnetic transition.

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