Magnetic-field and temperature dependencies of the electrical resistance near the magnetic and crystallographic first-order phase transition of $Gd_5(Si_2Ge_2)$

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The magnetic field (0 to 4 T) and temperature (5 to 320 K) dependencies of the electrical resistance of $Gd_5(Si_2Ge_2)$ have been measured. Upon heating in zero-magnetic field $Gd_5(Si_2Ge_2)$ undergoes a simultaneous magnetic and crystallographic phase transition at about 276 K. The electrical resistance of $Gd_5(Si_2Ge_2)$ changes drastically and has significant temperature and magnetic-field hystereses. The magnetoresistance has a negative peak of -26% between 274 and 295 K in a 4 T magnetic field, which is associated with the transition from the low-temperature, low-resistance ferromagnetic orthorhombic to the high-temperature, high-resistance paramagnetic monoclinic phase. The increase of the total resistance upon transformation from the magnetically ordered orthorhombic to magnetically disordered monoclinic phase correlates with the differences between the two crystallographic modifications of $Gd_5(Si_2Ge_2)$. The behavior of the electrical resistance as a function of magnetic field between 262 and 282 K shows the presence of temperature-dependent critical magnetic fields, which can reversibly transform both the magnetic and crystal structures of the material. The magnetic phase diagram obtained from the magnetic-field and temperature dependencies of the electrical resistance of $Gd_5(Si_2Ge_2)$ is proposed. [S0163-1829(99)06835-6]

INTRODUCTION

The giant magnetocaloric effect (GMCE) recently reported in the $Gd_5(Si_xGe_{1-x})_4$ alloys, where $0 \leqslant x \leqslant 0.5$,[1–3] warrants further experimental and theoretical studies of this class of ferromagnetic materials. The GMCE have been observed at the temperature where the ferromagnetic orthorhombic (FO) phase transforms into the high-temperature paramagnetic monoclinic (PM) phase.[3,4] In zero-magnetic field the Curie temperature, T_C, of the $Gd_5(Si_2Ge_2)$ alloy is 276 K and T_C increases with the increasing magnetic field, thus both magnetic and crystal structures of the $Gd_5(Si_2Ge_2)$ can be altered by a magnetic field.[1,4]

The $Gd_5(Si_2Ge_2)$ composition represents the silicon-rich boundary of the room-temperature monoclinic intermediate ternary phase $Gd_5(Si_xGe_{1-x})_4$, where $0.24 \leqslant x \leqslant 0.5$, which exists in the $Gd_5Si_4-Gd_5Ge_4$ pseudobinary system.[3] The temperature of the magnetic phase transition in $Gd_5(Si_xGe_{1-x})_4$ varies from 130 K for $x=0.24$ to 276 K for $x=0.5$.[3] It was shown[4] that the magnetic ordering in $Gd_5(Si_{1.8}Ge_{2.2})$ is accompanied by a structural (crystallographic) transition and that the crystallographic transition, therefore, could also be induced by the application or removal of a magnetic field. Both magnetic and structural transformations can lead to the changes in the charge carrier concentration and in the scattering mechanisms, which can be detected from the temperature and magnetic-field dependencies of the electrical resistance. The variation of the charge carrier concentration and the scattering mechanisms with both temperature and magnetic field have been studied in many intermetallic compounds based on the $4f$ and $5f$ elements.[5] In this paper we report on the experimental results of the temperature (5 to 320 K) and magnetic-field (0 to 4 T) dependencies of the electrical resistance of three different samples of the $Gd_5(Si_2Ge_2)$ alloy.

EXPERIMENTAL DETAILS

The alloy with a nominal composition of $Gd_5(Si_2Ge_2)$ was prepared by arc melting of the pure elements as described earlier.[1–3] Weight losses during arc melting were negligible, and therefore, the alloy composition was assumed unchanged. No impurity phases were detected by x-ray powder diffraction and optical metallography. Three different samples were cut from different locations in the original alloy, which had been cast into a finger. Sample #1 was cut from the middle part of a finger that had been heat treated for 1 h at 1300 °C. One half of the remaining finger was then heat treated for an additional 1 h at 1400 °C and samples #2 and #3 were cut from each end of this half of the finger, with sample #2 being near the middle of the original uncut finger. Samples for the transport measurements were cut using low speed diamond saw and had typical dimensions of $2 \times 2 \times 5$ mm.[3] Electrical connections to the samples were made by attaching thin platinum wires using H20E Epotek paste manufactured by Epoxy Technology. The dc electrical resistance measurements were carried out using a Lake Shore Model No. 7225 magnetometer equipped with a probe for making four-point electrical resistance measurements. The measurements were performed at constant dc current of 10 mA in the temperature range from 5 to 320 K and in magnetic fields from 0 to 4 T with the current applied in different directions to eliminate possible thermals. The magnetic-field vector **B** was oriented parallel to the direction of electrical

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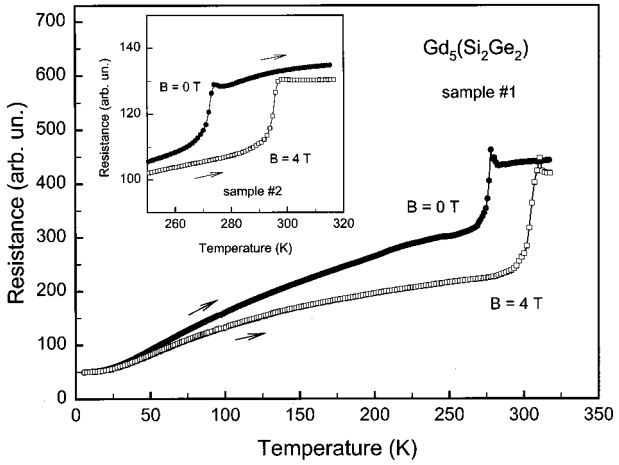

FIG. 1. Temperature dependence of the electrical resistance of the $Gd_5(Si_2Ge_2)$ sample #1 in zero and 4 T magnetic field. The inset shows the same for the $Gd_5(Si_2Ge_2)$ sample #2 near the Curie temperature.

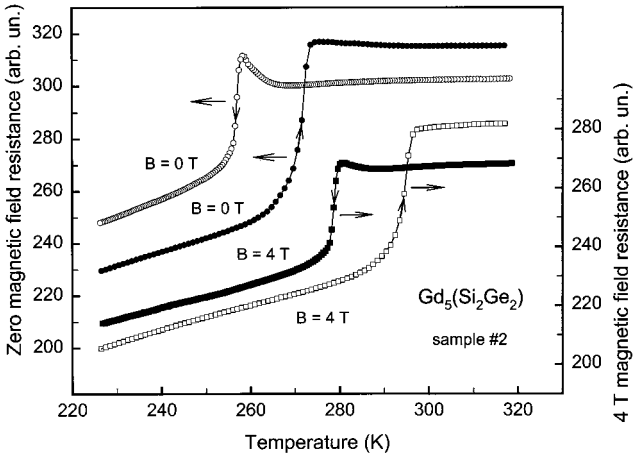

FIG. 2. Temperature dependence of the electrical resistance of the $Gd_5(Si_2Ge_2)$ sample #2 on cooling and heating in zero and 4 T magnetic fields.

current **j**. The error of resistance measurements was approximately 1%.

RESULTS AND DISCUSSION

The temperature dependence of the electrical resistance $R(T)$ for sample #1 measured during warming is presented in Fig. 1. In zero magnetic field the $R(T)$ dependence has a normal metallic character, but near $T_{cr} \approx 280$ K it shows a large increase of about 25% of its value before the transition, and after a small peak the resistance above ~280 K becomes weakly dependent on temperature. At 300 K the resistivity $\rho_{300\,K}$ of sample #1 is 2000 $\mu\Omega\cdot$cm and the residual resistance ratio (RRR) $\rho_{300\,K}/\rho_{5.5\,K} = 8.8$ (also see Table I). For sample #2, shown in the inset to Fig. 1, the temperature dependence of the resistance in the zero-magnetic field and the RRR ($\rho_{300\,K}/\rho_{5.5\,K} = 8.9$) are similar to that of sample #1, but the room-temperature resistivity is significantly smaller, $\rho_{300\,K} = 1100\,\mu\Omega\cdot$cm. The resistance for sample #2 increases by ~20% at $T_{cr} \approx 272$ K, which is slightly lower than that for sample #1. When measured in a 4 T magnetic field the resistance of sample #1 shows a large increase at $T_{cr} \approx 310$ K totaling about 100% of its value before the transition. For sample #2 the increase at $T_{cr} \approx 296$ K in 4 T is still 20%. For sample #3 the values of T_{cr} and the amplitude of the resistance discontinuity are similar to those of sample #2 (see Table I), but its room-temperature resistivity, $\rho_{300\,K} = 2800\,\mu\Omega\cdot$cm, is considerably larger. Application of a 4 T magnetic field shifts the critical temperature, T_{cr}, by 24 K (a rate 6 K/T), 26 K (a rate 6.5 K/T), and 30 K (a rate 7.5 K/T) for the samples #2, #3, and #1, respectively. Thus, the value of T_{cr} and its magnetic-field dependence are practically independent of the resistivity of the different samples and their heat treatment history. Slight differences in the values of T_{cr} could be due to the fact that $R(T)$ measurements are performed at a constantly changing temperature inside the measurement chamber back filled with a small amount of the helium heat exchange gas. The recorded temperature is actually the average temperature reading of two sensors mounted on the side of the measurement chamber above and below the sample. Therefore, since no temperature sensor is in direct contact with the sample, it is possible that the actual temperature of the sample is slightly different than the recorded reading. Furthermore, the possible difference between the heating (cooling) rates and the pressure of the helium heat exchange gas will also introduce slight variations of the actual sample temperature with respect to the recorded average temperature reading of the two temperature sensors. We estimate that both temperature uniformity and stability are within ± 2 K near room temperature.

In Fig. 2 we present the temperature dependence of the resistance for sample #2 for a cooling and heating cycle in zero and 4 T magnetic fields. A significant temperature hysteresis of the electrical resistance is observed in both cases. In the zero-magnetic field a value of a temperature hysteresis for sample #2 is 14 K. The temperature hysteresis of ~15 K is also preserved in a 4 T magnetic field. If one considers the

TABLE I. The electrical resistance of three different $Gd_5(Gi_2Ge_2)$ samples at 300 K, the resistance ratio, and the transition temperatures on cooling and heating at 0-T magnetic field and at 4 T.

			$T_{cr}(K)$			
			Cooling		Heating	
Sample	$\rho_{300\,K}$ $\mu\Omega\cdot$cm	$\rho_{300\,K}/\rho_{5.5\,K}$ $B=0$	$B=0$	$B=4$ T	$B=0$	$B=4$ T
#1	2000	8.8	-	-	280	310
#2	1100	8.9	258	281	272	296
#3	2800	8.9	259	281	273	299

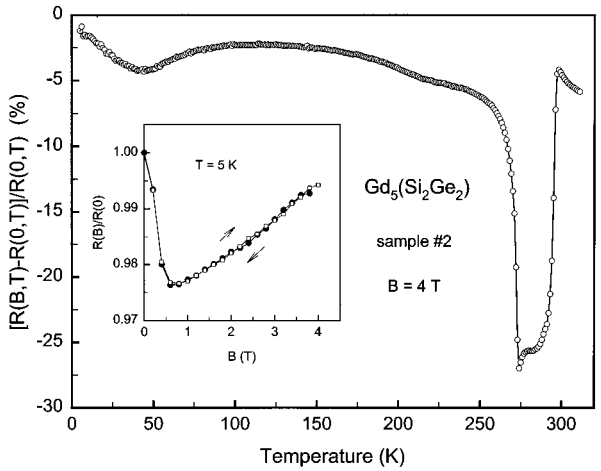

FIG. 3. Temperature dependence of the magnetoresistance of $Gd_5(Si_2Ge_2)$ sample #2 on heating in 4 T magnetic field. The inset shows the dependence of the electrical resistance $R(B)$ vs magnetic field at 5 K normalized to its zero-field value $R(0)$.

temperature hysteresis between the sample cooled in the zero-magnetic field and the sample heated in the 4 T magnetic field, its value reaches ~36 K. For the sample #3 the character of hysteresis in a zero and 4 T magnetic field is similar to the sample #2 (see Table I). For the sample #1 the temperature cycling measurements were impossible due to a large continuous increase of its resistivity during cycling. The most likely reason for this is an increase in the number of cracks in the sample during the cycling through the simultaneous magnetic and crystallographic transition (the RRR remains unchanged indicating the presence of temperature independent resistance due to microcracks). The resistance during cycling of samples #2 and #3 also increases gradually but much slower compared to sample #1 and maintains practically constant RRR. Thus, the samples, which were heat treated longer (1 h at 1300 °C plus 1 h at 1400 °C) have a more stable microstructure.

The behavior of the electrical resistance in the $Gd_5(Si_2Ge_2)$ samples could be understood as the result of transitions between the low resistance FO and the high-resistance PM phases during increasing or decreasing temperature. In zero-magnetic field both FO and PM phases coexist between 258 and 272 K in the $Gd_5(Si_2Ge_2)$ alloy. The value of the temperature hysteresis in the zero-magnetic field (14 K) is most likely controlled by the kinetics of the FO↔PM phase transition in the bulk alloy. Application of a magnetic field restrains the crystallographic transitions via the increase of the ferromagnetic ordering temperature, which leads to a magnetic field induced hysteresis of 36 K, of which about 22 K of the increase is due to the 4 T magnetic field change.

The temperature dependence of the magnetoresistance $[R(B,T)-R(0,T)]/R(0,T)$ in a 4 T magnetic field for sample #2, normalized to −1% at 5 K (see below) to exclude the contribution from microcracks, is presented in Fig. 3. It shows two anomalies: a small minimum near 44 K (−5%) and a pronounced sharp minimum between 274 and 295 K (−26%). The high-temperature minimum is associated with the first-order phase transition and its character is determined by the differences in the resistivity of the orthorhombic and monoclinic phases in the $Gd_5(Si_2Ge_2)$ sample. The nature of the low-temperature minimum is not clear, but it is possible, that it is due to partially antiferromagnetic interactions in the gadolinium sublattices. The inset in Fig. 3 shows the magnetic-field dependence of the resistance $R(B)$ at 5 K normalized to its zero-field value $R(0)$. The $R(B)/R(0)$ dependence is typical for ferromagnetic metallic materials. Consistent with the magnetic-field dependence of the magnetic moment at 5 K, the $R(B)/R(0)$ reduction (i.e., the negative magnetoresistance) below 0.7 T is associated with reaching saturation magnetization in the $Gd_5(Si_2Ge_2)$. Above 0.7 T the $R(B)/R(0)$ increases and it is typical for the magnetoresistance in a normal metal due to the Lorentz forces, which also indicates a simple electronic band structure for $Gd_5(Si_2Ge_2)$ near the Fermi level at low temperature. In magnetic fields varying from 0 to 4 T the low temperature electrical resistance of the $Gd_5(Si_2Ge_2)$, as well as its magnetization, shows nonhysteretic behavior (inset in Fig. 3).

At higher temperatures the magnetic-field dependence of the $Gd_5(Si_2Ge_2)$ resistance shows hysteresis, with the hysteretic behavior strongly dependent on a temperature interval. Figure 4 shows the $R(B)_{T=\text{const}}$ dependence in the three different temperature ranges: from 250 to 264 K [Fig. 4(a)] from 266 to 286 K [Fig. 4(b)] and from 288 to 308 K [Fig. 4(c)]. Below 262 K the $R(B)_{T=\text{const}}$ dependence displays a narrow hysteresis, but the values of the resistance at $B=0$ before the magnetic field was increased to $B=4\,\text{T}(R')$ and the resistance at the same temperature when the magnetic field was reduced back to $0\,\text{T}(R'')$ are the same. At 262 K and above the magnetic-field dependence of the resistance shows a different character when the magnetic field is cycled from 0 to 4 T and back to 0 T [Figs. 4(a), (b)]. First, at a temperature $262 \leqslant T \leqslant 286\,\text{K}$ the zero-magnetic field resistance is not restored after the field sweep, i.e., R' no longer equals to R''. Then between 266 and 282 K [see Fig. 4(b)] a rising magnetic field brings about a sharp decrease in the resistance beginning at various specific critical magnetic fields, B_{cr1}, and ending at $B_{\text{cr2}} > B_{\text{cr1}}$ depending upon the temperature.

The reason for this resistance drop can be understood if one assumes that even at temperatures slightly below the Curie temperature ($T_C \approx 276\,\text{K}$), a small fraction of the sample has already transformed to the high-resistance PM phase. The appearance of a small amount of PM phase below the usually observed T_C is possible because holding a sample even at temperature close but below the Curie temperature, allows the high-temperature phase to nucleate due to fluctuations of magnetic moment and temperature. The transformation in bulk, however, does not occur at this temperature due to insufficient superheating and/or for kinetic reasons. What is detected as the Curie temperature in the $Gd_5(Si_2Ge_2)$ is actually the temperature at which both nucleation and growth of the PM phase rapidly proceed through the bulk of material. Application of the magnetic field then reverses the transformation of this fraction of the paramagnetic phase to the low-resistance ordered ferromagnetic phase (just as would occur by lowering the temperature), which is also associated with the crystallographic transformation from the monoclinic to the orthorhombic structure causing a steplike drop in the resistance. During a magnetic-field decrease from 4 to 0 T the inverse FO→PM phase transition begins at different

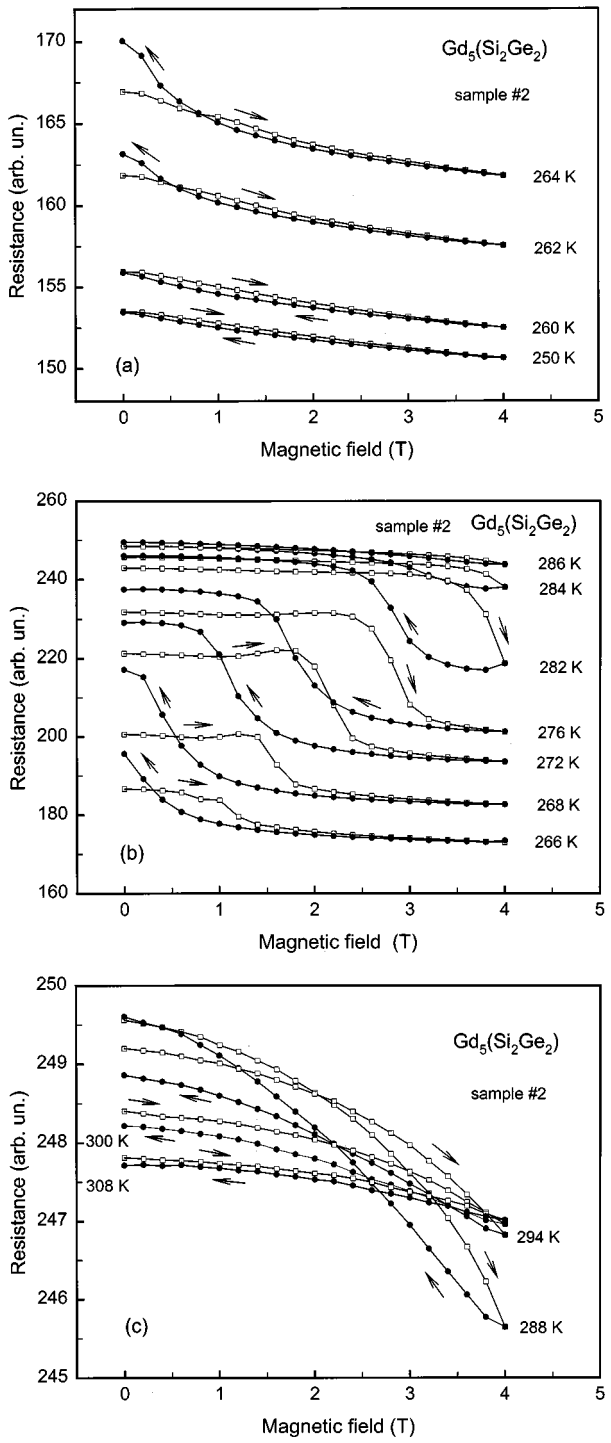

FIG. 4. Magnetic-field dependence of the electrical resistance of $Gd_5(Si_2Ge_2)$ sample #2 for the temperature intervals 250 to 264 K (a), 266 to 286 K (b) and 288 to 308 K (c).

specific critical fields, B_{cr3}, ending at $B_{cr4}<B_{cr3}$ which are lower than isothermal critical fields B_{cr1} and B_{cr2} (i.e., $B_{cr4}<B_{cr3}<B_{cr1}<B_{cr2}$). The temperature dependence of the values of all critical fields is shown in Fig. 5. They increase nearly linearly with temperature at a rate of 0.14 T/K and outline the ferromagnetic orthorhombic and the paramagnetic monoclinic phase fields together with the two phase regions for an isothermal change of the magnetic field. Extrapolation of the described behavior to zero-magnetic

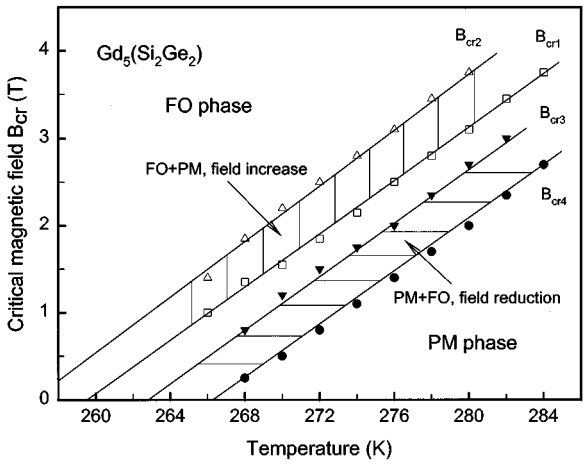

FIG. 5. Critical magnetic fields for $Gd_5(Si_2Ge_2)$ sample #2 during an isothermal increase (B_{cr1},B_{cr2}) and decrease (B_{cr3},B_{cr4}) of the magnetic field.

field indicates that at the FO→PM phase transition the high-temperature phase could nucleate at a temperature as low as ~263 K, while upon the PM→FO transition the low-temperature phase could nucleate at ~260 K. The full isothermal magnetic-field hysteresis for the FO↔PM phase transition is ~1.7 T and the isofield temperature gap between B_{cr2} and B_{cr4} is approximately 10 K. These results for the $Gd_5(Si_2Ge_2)$ samples are similar to the results reported based on the magnetostriction data for the $Gd_5(Si_{1.8}Ge_{2.2})$ sample.[4] In the latter case critical magnetic fields increases at the rate of ~0.2 T/K and the observed temperature gap is ~7 K, both of which are similar to what we have measured on $Gd_5(Si_2Ge_2)$, i.e., ~0.14 T/K and ~10 K, respectively.

One of the interesting experimental results obtained in this study is the inequality of the zero-field resistance after the magnetic field sweep, i.e., the fact that $R''>R'$ at any temperature between 262 and 286 K [Figs. 4(a) and 4(b)]. Two possible reasons for this are as follows: (1) in the $Gd_5(Si_2Ge_2)$ not all of the high-resistance paramagnetic monoclinic phase transforms back into the low-resistance ferromagnetic orthorhombic phase after removal of the magnetic field, and (2) when the FO→PM phase transition occurs the volume expansion, which in the case of $Gd_5(Si_{1.8}Ge_{2.2})$ is approximately 0.4%,[4] promotes development of microcracks in the specimen. The same takes place for the inverse PM→FO phase transition. It is difficult to single out one of the two likely mechanisms without the studies of the kinetics of the first-order phase transition in $Gd_5(Si_2Ge_2)$, and it is feasible that both contribute to the discussed phenomenon. The difference between the zero-magnetic field resistance is temperature dependent and the ratio $\Delta=(R''-R')/R'$ has a maximum at 268 K. In general, our results for the electrical resistance of $Gd_5(Si_2Ge_2)$ are similar to the experimental results for $Gd_5(Si_{1.8}Ge_{2.2})$ (Ref. 6) and this indicates a similarity in the charge carrier scattering mechanisms in these two materials.

The temperature dependence of an electrical resistivity $\rho(T)$ in rare-earth-based intermetallic compounds, such as $Gd_5(Si_2Ge_2)$ in the zero-magnetic field has the following main components:

$$\rho(T)=\rho_0+\rho_{\text{el-ph}}(T)+\rho_{\text{el-el}}(T)+\rho_{\text{el-mag}}(T), \qquad (1)$$

where ρ_0 is the residual temperature independent resistivity, $\rho_{\text{el-ph}}(T)$ is the resistivity due to the electron-phonon scattering, $\rho_{\text{el-el}}(T)$ is the resistivity due to electron-electron scattering, and $\rho_{\text{el-mag}}(T)$ is the resistivity due to electron-magnon interactions. For the $Gd_5(Si_2Ge_2)$ alloy it is important to recognize that the temperature independent resistivity due to the presence of microcracks, ρ_{crack}, contributes to the ρ_0 component and it can change upon cycling the material through the phase transition due to a discontinuous changes in the lattice parameters. It appears that the large value of the ρ_0 in $Gd_5(Si_2Ge_2)$ alloy (110–310 $\mu\Omega$ cm) is caused by the presence of cracks.

In the region of the first-order magnetic and crystallographic phase transition the two most important contributions to the electrical resistivity of $Gd_5(Si_2Ge_2)$ become the $\rho_{\text{el-ph}}$ and $\rho_{\text{el-mag}}$ components. In most ferromagnetic materials the $\rho_{\text{el-mag}}$ component usually approaches maximum at the T_C and becomes weakly dependent on the temperature above the T_C. The disappearance of the temperature dependence of the electron-magnon scattering correlates well with the reduction of dR/dT value above the T_C (Figs. 1 and 2). However, our data also indicate that the resistivity of the paramagnetic monoclinic phase in $Gd_5(Si_2Ge_2)$ is larger by at least 20% compared with that of the ferromagnetic orthorhombic phase. It means that the combined electron-phonon and electron-magnon scattering in the monoclinic phase are much stronger than those in the orthorhombic phase. Alternatively one could assign the increased resistance to a decrease of the charge carrier concentration after FO to PM phase transition. We assume, however, that the change in the electron-phonon scattering is the primary mechanism responsible for the increased resistance in the paramagnetic monoclinic phase. Furthermore, the complete study of the crystal structures of the low-temperature and the high-temperature $Gd_5(Si_2Ge_2)$ phases[7] indicates that the major crystallographic change occurs due to the breakup of $\frac{1}{2}$ of covalentlike Si-Si, Si-Ge, and Ge-Ge bonds upon the transition from the low-temperature orthorhombic to the high-temperature monoclinic phase. This structural transition is indeed quite similar to the transition, which occurs upon the change of chemical composition from $Gd_5(Si_{2+\delta}Ge_{2-\delta})$ to $Gd_5(Si_2Ge_2)$, where the former has room-temperature orthorhombic, while the latter has room temperature monoclinic crystal structure and is also accompanied by the similar changes in the atomic bonding.[3] The monoclinic lattice, therefore, is loosely bonded and more disordered compared to the orthorhombic one, which in turn results in the increased electron-phonon scattering leading to the increase in the overall resistivity of the $Gd_5(Si_2Ge_2)$ alloy above the T_c.

CONCLUSION

The anomalous behavior of the magnetoresistance has been observed in a variety of $4f$ and $5f$ elements-based intermetallic compounds. In some of them the resistivity changes with magnetic field because of the presence of a metamagnetic transition.[5,8] Since the negative magnetoresistance is fundamentally limited to -100%, a -20% or larger absolute resistance change associated with metamagnetism is sometimes referred to as the giant magnetoresistance (GMR) following the analogy of the bulk rare-earth intermetallic materials with layered crystal structures to the thin-film multilayered systems with the GMR.[8,9] It is well known that the giant magnetoresistance can be observed in rare-earth materials other than in materials that exhibit a metamagnetic transition. For example, in the $CeMn_{2-x}Si_{2+x}$ system with a Mn sublattice in the antiferromagnetic state and Ce in an intermediate valence state it was found that the positive magnetoresistance reaches about 230% at 4.2 K in a magnetic field of 13 T.[10] The nature of the large magnetoresistance in both $Gd_5(Si_2Ge_2)$ and $Gd_5(Si_{1.8}Ge_{2.2})$,[6] however, is not similar to the nature of the GMR in the multilayered systems. The negative magnetoresistance in the $Gd_5(Si_xGe_{1-x})_4$-type alloys ($x=0.5$, 0.45) with a first order magnetic and crystallographic phase transition is determined by the difference in the electron-phonon and electron-magnon scattering of the charge carriers in the low-temperature ferromagnetic orthorhombic phase and the high-temperature paramagnetic monoclinic phase. It may become positive if the high-temperature phase has lower resistance compared to the low-temperature phase. The hysteretic behavior of the electrical resistance in zero and nonzero magnetic fields in the region of the first-order phase transition is associated with the kinetics of the phase transformation process and needs further study.

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