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Hall effect in $Gd_5(Si_{1.8}Ge_{2.2})$

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We have measured the Hall effect of polycrystalline $Gd_5(Si_{1.8}Ge_{2.2})$ in the temperature range 4–360 K and in magnetic fields of up to 12 T. The Hall resistivity follows the magnetization of the material in the range studied. The anomalous Hall coefficient increases with increasing temperature and shows a sharp dip at the magnetostructural transition. Away from the dip, the Hall coefficient scales roughly with the square of the total resistivity, as expected if side-jump scattering dominates.

Recently, there has been much interest in a $Gd_5(Si_xGe_{1-x})_4$ system with $x \lesssim 0.5$. This system exhibits a ‘‘giant’’ magnetocaloric effect.[1,2] Alloys with $0.24 \leqslant x \leqslant 0.5$ that crystallize in the monoclinic structure at room temperature are of special interest. Upon cooling, these alloys undergo a first-order transition from a paramagnetic to a ferromagnetic phase. The Curie temperature T_c varies linearly with composition, from 276 K for $x=0.5$ to approximately 140 K for $x=0.24$. This transition is accompanied by a large magnetic entropy change, which has been claimed to be the largest value ever reported for any magnetic solid.[1] However, direct adiabatic temperature change measurements show a significantly smaller magnetocaloric effect and place it in the same range as that of Gd and its alloys.[3] Nevertheless, $Gd_5(Si_xGe_{1-x})_4$ alloys are attractive for magnetic refrigeration technology as the first-order magnetic transition is reversible with respect to an alternating magnetic field.

It has been reported quite recently that the magnetic transition in $Gd_5(Si_{1.8}Ge_{2.2})$ is associated with a first-order structural transition from a monoclinic (paramagnetic) to an orthorhombic (ferromagnetic) structure.[4] This magnetostructural transition is hysteretic and can be driven reversibly by applying an external magnetic field. Strong magnetoelastic effects are observed at T_c.[4] There is also a large negative magnetoresistance of about 20% in this temperature region.[5] Comparable negative magnetoresistance values have been found in $Gd_5(Si_2Ge_2)$ near the transition from the orthorhombic ferromagnetic low-temperature phase to the monoclinic paramagnetic high-temperature phase.[6] However, we could find no additional reports on the electronic properties of $Gd_5(Si_xGe_{1-x})_4$ alloys even though their physical properties as well as their applications seem to be very interesting. Recently, large variations of the Hall effect in the vicinity of magnetic phase transition points have been observed.[7] Consequently, this effect may be a sensitive probe of critical behavior. In addition, the Hall effect gives information about the mechanism behind the scattering of carriers. In this paper we report results of Hall-effect measurements in $Gd_5(Si_{1.8}Ge_{2.2})$ polycrystalline samples as a function of temperature in the range 4–360 K and of magnetic field up to 12 T. The Hall resistivity follows the magnetization of the material in the range studied. The anomalous low-field Hall coefficient R_s shows a sharp dip at the Curie temperature that we relate to the magnetostructural character of the transition. Away from the dip, R_s scales roughly with the square of the total resistivity as expected when side-jump scattering dominates.

The $Gd_5(Si_{1.8}Ge_{2.2})$ alloy was synthesized by arc melting. We have already studied its magnetostructural and magnetoresistance properties. Details on the material preparation and its characterization can be found elsewhere.[4] The Hall-effect measurements were performed on bar-shaped samples with the six-probe method. Contact leads were ultrasonically soldered to the samples. The magnetization as a function of temperature was measured using a commercial superconducting quantum interference device magnetometer. Magnetization measurements were performed on the same samples that were used in magnetotransport studies. Isotherms of both magnetization and Hall resistivity were measured in a superconducting coil producing steady magnetic fields of up to 12 T.

Figure 1 shows how the magnetization M (upper panel) and the Hall resistivity ρ_H (lower panel) of $Gd_5(Si_{1.8}Ge_{2.2})$ vary with magnetic field at three different temperatures. It is clear that the ρ_H data, which are electronlike, follow the magnetization of the sample. The low-field Hall resistivity is shown in Fig. 2. It increases with increasing temperature up to ≈ 235 K where it drops abruptly; ρ_H decreases slowly thereafter. Phenomenologically, the Hall resistivity is given by $\rho_H = R_o \mathbf{B} + R_s 4\pi \mathbf{M}$, where R_o is the ordinary Hall coefficient, $\mathbf{B}$ is the applied magnetic induction, and $\mathbf{M}$ is the spontaneous magnetization. In our samples, $R_o \ll R_s$ as can be ascertained from the high-field behavior, where ρ_H is flat. Consequently, we obtain the values of the anomalous (or spontaneous) Hall effect coefficients from $R_s = \rho_H / 4\pi M$, where ρ_H and M are the low-field Hall resistivity and magnetization, respectively, measured with the same field (0.1 T) applied to the same sample. The temperature variation of magnetization is shown in the left-side inset of Fig. 2, both for cooling and warming conditions. M also drops sharply at T_c. There is a small thermal hysteresis of about 5 K in both M and ρ_H. Figure 3 shows how R_s varies with temperature in $Gd_5(Si_{1.8}Ge_{2.2})$ upon cooling. A sharp minimum is seen at the transition temperature on the curve that otherwise increases monotonically with temperature. There is also a small dip at about 280 K, which is expected to arise from a minor secondary orthorhombic phase $[Gd_5(Si_{2.1}Ge_{1.9})]$ present in the sample and which orders ferromagnetically at

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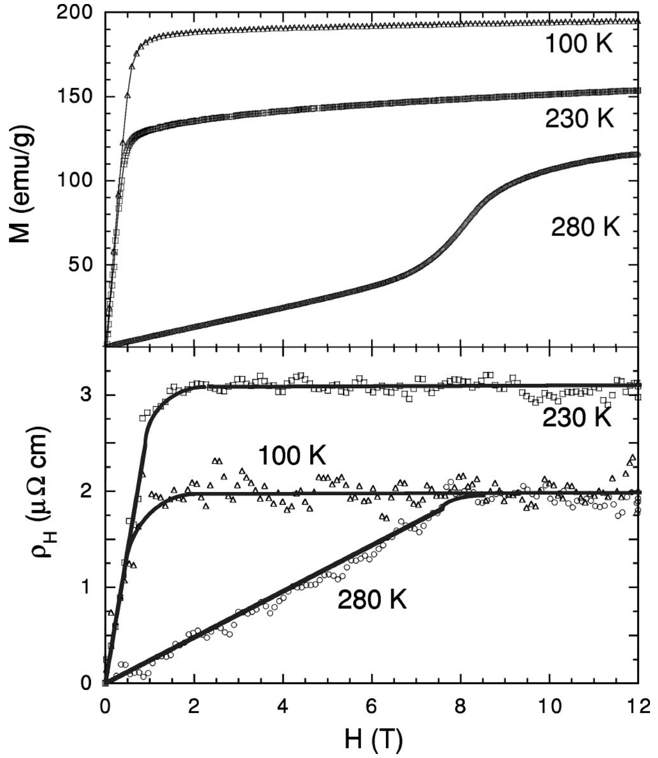

FIG. 1. Magnetization (upper panel) and Hall resistivity (lower panel) as a function of magnetic field in $Gd_5(Si_{1.8}Ge_{2.2})$ at three different temperatures. Solid lines are a guide to the eye. Data points for $T = 280$ K correspond to a decreasing magnetic field.

this temperature.[4] The sharp minimum in $R_s(T)$ curve shows hysteretic behavior through the transition; it is also larger upon warming as shown in the upper inset of Fig. 3.

A simple relation is usually satisfied between the anomalous Hall coefficient and the longitudinal resistivity ρ: $R_s = a\rho + b\rho^2$, where the first term stands for a skew component,[8] and the second term gives the side-jump contribution.[9] Both terms are brought about by spin-orbit coupling. Dilute impurities and spin disorder are expected to give $R_s \propto \rho$, whereas concentrated defects and phonons lead to $R_s \propto \rho^2$. In our samples, the value of the residual resistivity is relatively high (73 $\mu\Omega$ cm), likely arising from the high concentration of impurities and or static defects. Therefore, the latter term is expected to dominate. This is shown in the lower inset of Fig. 3, where the solid line exhibits ρ^2 fit to the data. Here, the data points below the magnetostructural transition are shown. The best fit to $R_s = b\rho^2$ yields $b = 0.0014\ \Omega^{-1}\ \mathrm{cm}^{-1}\ \mathrm{G}^{-1}$. Such a value is predicted by the theory for one-band ferromagnetic materials assuming electron concentration of about $2\times10^{22}\ \mathrm{cm}^{-3}$ and a constant lateral displacement of the charge carrier's trajectory at every scattering of 7×10^{-10} cm.[9] Therefore, side-jump scattering explains well our experimental results away from the magnetostructural transition region. It is more difficult to interpret the sharp minimum in R_s observed at this transition. Kondo and Maranzana, considering the spin-orbit interaction between localized spin and the current carriers, have shown that skew scattering is proportional to the third moment of the magnetization fluctuations.[10,11] Spin fluctuations are expected to be large near critical points. However, this is not the case here since the magnetic transition at T_c is of first order. On the other hand, a change of carrier concentration through the transition may explain the observed anomaly. Such a scenario seems plausible since the transition at T_c is not only magnetic but also a structural one from a monoclinic to an orthorhombic phase. This would lead to a different density of states at the Fermi level and, consequently, to a variation of the Hall resistivity. However, the inspection of the right-side inset of Fig. 2, which exhibits how the normalized Hall resistivity changes with temperature at different magnetic fields, suggests that strong spin scattering rather than the electron density is responsible for the large variation of R_s at the Curie temperature. It is clear that an increasing magnetic field smears out the drop in ρ_H at the transition and moves it to higher temperatures. This is consistent with results of magnetoresistance and magnetostriction measurements.[4–6] Thus, at present it is difficult to identify the mechanism underlying the observed anomaly in R_s.

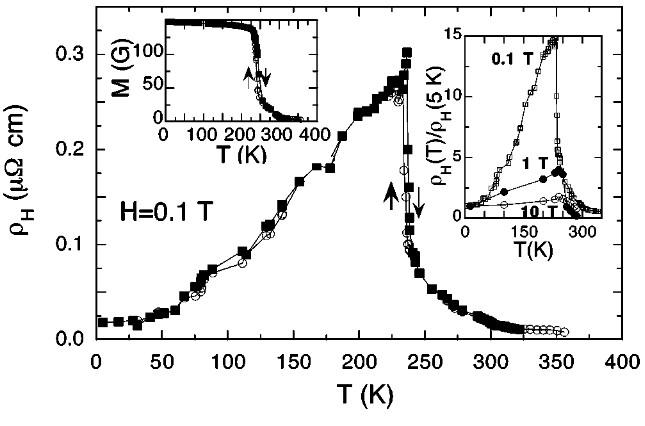

FIG. 2. Low-field Hall resistivity as a function of temperature upon warming (full squares) and cooling (open circles) in $Gd_5(Si_{1.8}Ge_{2.2})$. The magnetization measured at the same field is shown in the inset on the left-hand side. Normalized Hall resistivity as a function of temperature at three different magnetic fields is exhibited in the right-hand side inset. Solid lines are a guide to the eye.

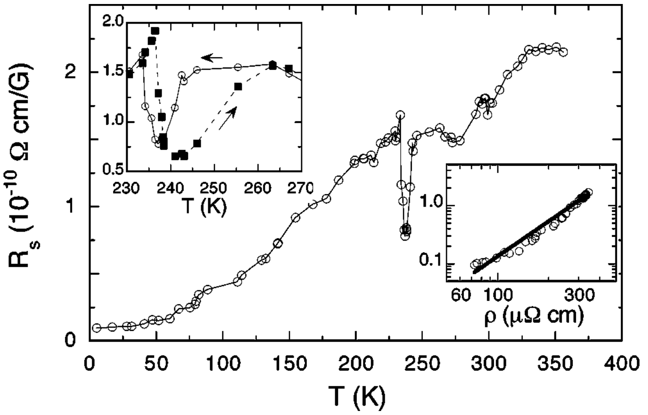

FIG. 3. Anomalous Hall coefficient $R_s = \rho_H/4\pi M$ as a function of temperature for $Gd_5(Si_{1.8}Ge_{2.2})$. The upper inset shows $R_s(T)$ near the transition at warming and cooling conditions. Solid and dashed lines are a guide to the eye. R_s as a function of the total resistivity is exhibited in the lower inset. Here, the solid line shows the ρ^2 fit to the experimental data.

Finally, we would like to make a comment on the behavior of the anomalous Hall coefficient for temperatures above the magnetostructural transition. The theory predicts that ρ_H is proportional to the magnetic susceptibility in the paramagnetic region.[12] However, our measurements, which were made up to 360 K, correspond mainly to the metamagnetic region above the transition (240–310 K). R_s slowly increases in this region and starts to decrease at about 350 K. Thus, we do not reach a completely paramagnetic region in our measurements.

To summarize, we have studied the anomalous Hall effect in $Gd_5(Si_{1.8}Ge_{2.2})$. The anomalous Hall coefficient was found to scale as a square of the longitudinal resistivity. Such dependence can be produced by side-jump scattering. The anomaly in R_s, observed at the first-order transition from the low resistivity ferromagnetic orthorhombic phase to the higher resistivity paramagnetic monoclinic phase, is unexplained at present. Strong spin scattering as well as changes in the Fermi surface that take place through the transition may contribute to this phenomenon.

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