EFFECT OF THE ANTIFERROMAGNETIC-FERROMAGNETIC TRANSITION ON THE GALVANOMAGNETIC PROPERTIES OF $Mn_{1.88}Cr_{0.12}Sb$

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Results of an experimental investigation of the temperature dependences of the electric resistance and thermoelectric power, and also of the temperature and field dependences of the magneto-resistance ratio $\Delta R/R$ and Hall effect, are presented. Measurements were performed on a single crystal of $Mn_{1.88}Cr_{0.12}Sb$ in the range 77—380°K, which includes the antiferromagnetic-ferromagnetic transition temperature T_S. The transition changes all the mentioned properties sharply. The result $dT_S/dH = (-0.33 \pm 0.04) \times 10^{-3}$ deg/Oe is derived from the shift of the maximum of $\Delta R/R$ with variation of the field. The different characters of the temperature and field dependences of the galvanomagnetic effects in ferromagnetic and antiferromagnetic regions of a single crystalline structure indicate that the magnetic structure of matter is an essential factor in these effects.

INTRODUCTION

COMPOUNDS having the general formula $Mn_{2-x}Cr_xSb$ undergo two magnetic phase transitions as their temperature is varied. While possessing antiferromagnetic properties at low temperatures, at a temperature T_S they are transformed into a ferromagnetic state, which is destroyed at a still higher point Θ_f.[1] Investigations of these compounds in strong pulsed magnetic fields have shown that their antiferromagnetic state can be destroyed by high fields above a threshold value H_n which depends on temperature.[2] It has been established by means of x-ray and neutron-diffraction studies[3] that the antiferromagnetic-ferromagnetic transition at T_S involves only a change in the magnetic structure of the compound without affecting the crystallographic symmetry of the lattice. The lattice possesses P4/nmm tetragonal symmetry both above and below the transition temperature. For this reason at least two lines of investigation of the aforesaid compounds are of interest.

First, since only the spin symmetry is altered in a transition without affecting the lattice symmetry, it becomes possible to investigate the influence of spin ordering on the electric and galvanomagnetic properties of the substances.

Second, the antiferromagnetic-ferromagnetic transition is of intrinsic interest. The nature of such transitions is still not well understood although they have been observed in many compounds, alloys, and a large group of rare-earth metals.

The present article contains data regarding the temperature dependences of electric resistivity and thermoelectric power, and also the temperature and field dependences of magnetization, the magneto-resistance ratio $\Delta R/R$, and the Hall effect in single crystals of $Mn_{1.88}Cr_{0.12}Sb$ for the temperature range 77—380°K, which includes the antiferromagnetic-ferromagnetic transition point $T_S = 319$°K.

PREPARATION OF SAMPLES AND MEASUREMENT PROCEDURE

Single crystals of $Mn_{1.88}Cr_{0.12}Sb$ were prepared as follows. Fine powders of chemically pure Mn, Cr, and Sb[1)] were thoroughly mixed and pressed into the form of 50 × 8 × 7 mm rods. The compressed rods were placed in quartz ampoules which were highly evacuated, filled with purified helium, sealed, and heated slowly in a furnace to 700°C, at which temperature they were maintained for six hours. The temperature was then increased to 1000—1050°C in order to melt the samples. Crystallization of the melt was brought about by withdrawing the ampoules from the furnace at the rate 0.4 cm/hr during a period of 10—11 hrs. X-ray and microstructural phase analyses of the alloys then revealed traces of MnSb in addition to the principal phase, $Mn_{1.88}Cr_{0.12}Sb$. The crystallographic directions in the single crystals were determined by the

[1)] Electrolytic manganese and chromium degassed in a vacuum were used. The antimony contained the following impurities: Pb — 1.5×10^{-3}%, As — under 3×10^{-3}%, Co and Ni — 6×10^{-4}%, and Bi — under 4×10^{-5}%.

Laue x-ray method and by the magnetic method employing a Cardan (universal) suspension.

The isotherms of magnetization were obtained using the Domenicali-type pendulum magnetometer,[4] in fields having the maximum 19 kOe.

The electric resistivity and its magnetic dependence ($\Delta R/R$) as well as the Hall effect were measured using a PPTN-1 potentiometer. The technique of the Hall-effect measurements has been described in[5]. These measurements were obtained for two magnetic field and current directions in order to exclude extraneous effects; the maximum magnetic field was 28 kOe. In the measurements of magnetization and the galvanomagnetic effects the tetragonal axis of the monocrystalline sample was parallel to the external magnetic field, so that for $T > T_S$ it coincided with the axis of easy magnetization.

The thermoelectric power was determined for $Mn_{1.88}Cr_{0.12}Sb$ against copper, with the thermal current flowing parallel to the tetragonal axis. The sign of the thermo-emf was determined by comparison with standard samples of bismuth and antimony.

RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the specific magnetization σ, the Hall emf per unit of current density $E_x d/i$ (where E_x is the measured Hall emf, d is the thickness of the sample, and i is the current), and the transverse magnetoresistance ratio $\Delta R/R$, measured in a 15-kOe field. The temperature dependence of the resistivity, $\rho(T)$, is also shown. All of the foregoing properties are seen to change sharply in the temperature region 300—330°K, where we find a sharp rise of magnetization and of the Hall emf, a maximum of the magnetoresistance, and a 24% reduction of electric resistivity. The given temperature range includes the point $T_S = 319$°K of the antiferromagnetic-ferromagnetic transition. (The procedure used to determine T_S will be discussed below.) It is noteworthy that a change in the character of spin ordering is accompanied by changes not only in the absolute values of the galvanomagnetic effects $\Delta R/R$ and the Hall emf, but also in their signs. In the antiferromagnetic low-temperature range $\Delta R/R$ and the Hall emf are positive, but their absolute values are considerably smaller than in the ferromagnetic state. For example, at 77°K and 15 kOe we have $\Delta R/R = 0.8 \times 10^{-5}$. Above T_S, where ferromagnetic spin ordering exists, $\Delta R/R$ and the Hall emf are negative.

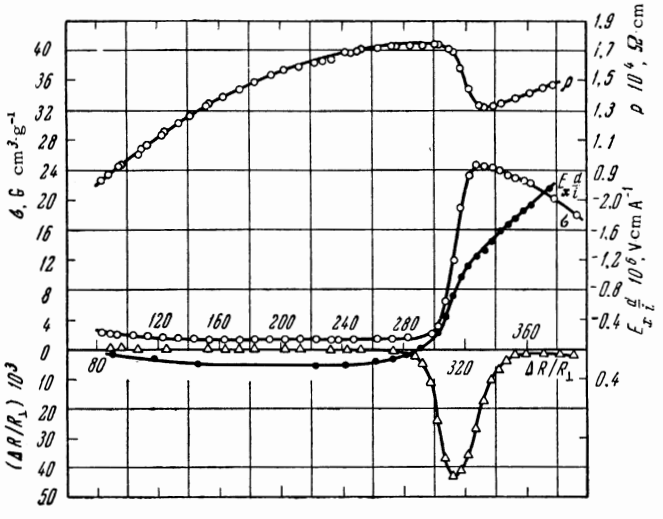

FIG. 1. Temperature dependences of the electric resistivity ρ, specific magnetization σ, Hall emf $E_x d/i$, and magnetoresistance ratio $\Delta R/R$.

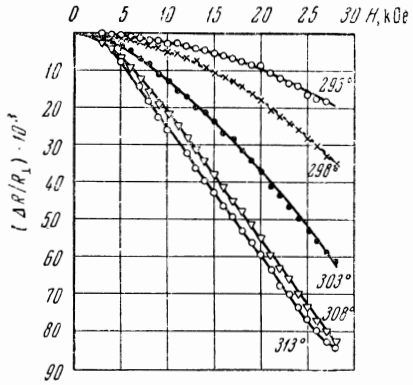

FIG. 2. Isotherms of the magnetoresistance ratio $\Delta R/R$ for $T < T_S$.

Figures 2 and 3 show isotherms of the transverse magnetoresistance ratio, measured both below (Fig. 2) and above (Fig. 3) the transition temperature. These measurements were used to plot the temperature dependence of the transverse

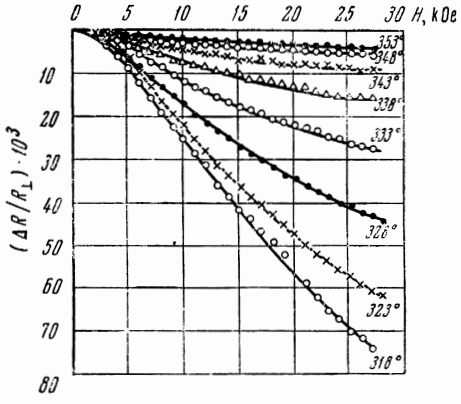

FIG. 3. Isotherms of the magnetoresistance ratio $\Delta R/R$ for $T > T_S$.

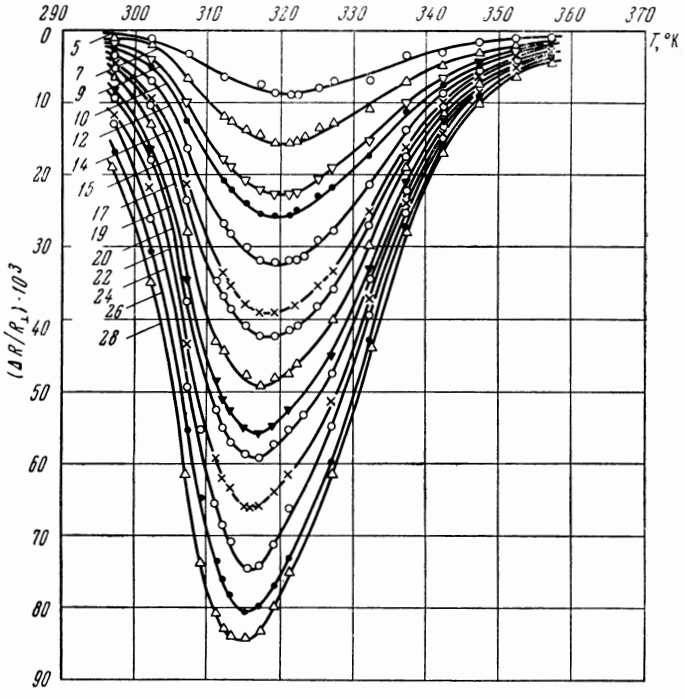

FIG. 4. Temperature dependences of the magnetoresistance ratio for different external magnetic fields.

magnetoresistance for different magnetic field strengths (Fig. 4). Here as for ordinary ferromagnets at the Curie point we observed the maximum of $\Delta R/R$ at the transition temperature, although the magnitude of the effect is almost twice as great as for ferromagnetic-paramagnetic transitions.[6] In the present case, moreover, the transition temperature T_S depends on the external magnetic field; with increase of the field the temperature corresponding to the maximum of $\Delta R/R$ is shifted. Figure 5 shows the variation of T_S determined from the maximum of the magnetoresistance ratio as a function of the magnetic field; it is seen that the antiferromagnetic-ferromagnetic transition temperature decreases as the field rises. Extrapolation of the straight line to zero field yields the true transition temperature $T_S = 319 \pm 0.5$°K. The variation of T_S with the magnetic field is

$$dT_s / dH = (-0.33 \pm 0.04) \cdot 10^{-3} \text{ deg/Oe}. \qquad (1)$$

It should be noted that the obtained value of dT_S/dH corresponds to the reciprocal of the value of

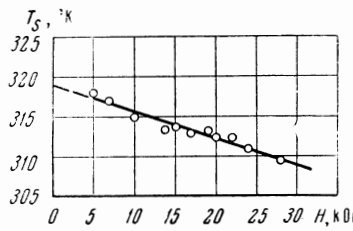

FIG. 5. Transition temperature T_S versus magnetic field.

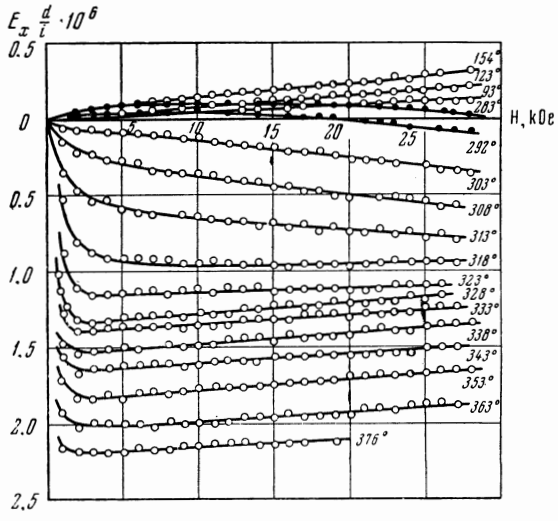

FIG. 6. Isotherms of the Hall emf.

dH_n/dT, the temperature variation of the threshold field H_n at which the antiferromagnetic state is destroyed.[2]

The Hall effect measurements are represented in Fig. 6, where it is seen that in the low-temperature antiferromagnetic region the Hall emf depends linearly on the external magnetic field only up to 154°K. In the range 154—280°K this linear dependence breaks down, while at 283° and 292°K the sign of the effect is reversed as the magnetic field increases. In the region of ferromagnetic ordering of the spins, i.e., for $T > T_S$, the magnetic dependence of the Hall potential difference is the same as for ordinary ferromagnets. The Hall field can then be represented as the sum of two terms, one of which is proportional to the field, while the other is proportional to the magnetization. When investigating the Hall effect in this temperature region ($T > T_S$) we were principally interested in the spontaneous Hall constant R_S, which was calculated from the relation $R_S = (E_S d/i)/\sigma_S$, where $E_S d/i$ is the spontaneous Hall emf that is determined by extrapolating to zero field the linear part of the $E_X d/i(H)$ curves for large fields. The spontaneous magnetization σ_S was taken to be the saturation magnetization of a sample at a given temperature. When the calculated values of R_S are examined we find a notably large value of the spontaneous Hall constant. For example, at $T/\Theta_f = 0.605$ for $Mn_{1.88}Cr_{0.12}Sb$ we have $R_S = 547 \times 10^{-10}$ V-g/A-G-cm^2, while for nickel we have $R_S = 6.8 \times 10^{-10}$ V-g/A-G-cm^2.[2)] Figure 7 shows that R_S

[2)]All data given in this article were obtained by measuring a single sample. Measurements performed on other samples cut from the same single crystal showed similar temperature and field dependences of all the investigated parameters, although the absolute values varied for the different samples as follows: $\rho \sim 30\%$, $\sigma \sim 10\%$, and $R_s \sim 18\%$, – apparently because of nonuniform chromium distribution in the alloy.

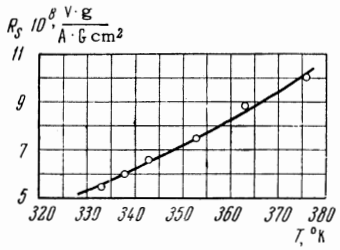

FIG. 7. Temperature dependence of the spontaneous Hall constant R_S.

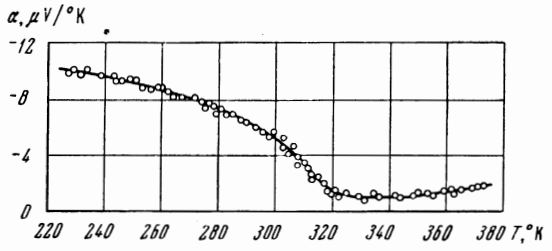

FIG. 8. Temperature dependence of the thermoelectric power.

for $Mn_{1.88}Cr_{0.12}Sb$ increases with the temperature. This accounts for the rise of the Hall emf in the region $T > T_S$, when the magnetization begins to diminish with rising temperature.

Figure 6 shows that the field constants R_S and R_0 in the Hall effect have different signs; R_S is negative, while the "ordinary" Hall constant R determined from the slope of $E_X(H)$ in high fields, is positive. It can thus be assumed that holes comprise the majority carriers in $Mn_{1.88}Cr_{0.12}Sb$. This agrees with recent work,[7] in which the Hall effect was measured only at room temperature in two samples of $Mn_{2-x}Cr_xSb$ having different chemical compositions ($x_1 = 0$ and $x_2 = 0.16$). Since the transition temperature depends on the chromium content, at the given temperature one sample was ferromagnetic, while the other was antiferromagnetic. From the Hall constant of the antiferromagnetic sample and the field constant R_0 of the ferromagnetic sample it was concluded that the sign and number of electric current carriers remains unchanged in these compounds when the character of spin ordering changes. However, our data indicate complicated temperature and field dependences of the Hall emf in $Mn_{2-x}Cr_xSb$ compounds. The sign of the effect depends on both temperature and the field; the slope of the $E_X(H)$ curves in the antiferromagnetic region changes with the temperature. Therefore measurements of the Hall effect at a single temperature, as in[7], do not yield unambiguous conclusions regarding the type and number of carriers when an antiferromagnetic-ferromagnetic transition occurs. Moreover, as already stated in the literature,[8,9] it is not always possible to determine R_0 from the slope of $E_X(H)$ in high magnetic fields when investigating the Hall effect in ferromagnets, because the "paraprocess" is then neglected.

In order to obtain independent information regarding the type of majority carriers we investigated the temperature dependence of the thermoelectric power α in $Mn_{1.88}Cr_{0.12}Sb$ within the range 220—380°K. The data, represented in Fig. 8, show that at the magnetic transition temperature the thermoelectric properties, like all other physical properties investigated in the present work, exhibit an anomaly that resembles the anomalous change of electric resistivity and is associated with the reduced thermoelectric power following an antiferromagnetic-ferromagnetic transition. The sign of the thermoelectric power is not positive, as would be expected from the determination of R_0, but is negative in the entire investigated temperature range, i.e., in both the ferromagnetic and antiferromagnetic states. In view of the foregoing discussion it can be assumed that the different signs of R_0 and α in the ferromagnetic state of the sample resulted from errors in determining R_0 because the paraprocess was not taken into account. However, it is difficult to account for the different signs of R_0 and α at low temperatures, i.e., in the antiferromagnetic state. Therefore we cannot arrive at a final conclusion regarding the majority carriers in this compound.

The considerable change of the thermoelectric power accompanying the transition indicates that a changed magnetic structure of the compound is accompanied by a changed concentration of carriers due to an altered degree of d-electron localization. This hypothesis is also supported by neutron-diffraction investigations,[3] which have established that the magnetic phase transition in $Mn_{2-x}Cr_xSb$ compounds changes the magnitudes of the magnetic moments MnI and MnII occupying different crystallographic locations. In the antiferromagnetic state ($T < T_S$) the magnetic moments MnI and MnII are 1.4 and 2.8 μ_B, respectively. With the ferromagnetic transition (for $T > T_S$) MnI increases by 0.4 μ_B, while MnII decreases by 0.5 μ_B. Therefore the increased electric conductivity in the ferromagnetic state can result not only from greater mobility (because of less scattering on ferromagnons than on antiferromagnons), but also from an increased number of carriers, affecting such kinetic phenomena as the electric conductivity and thermoelectric power.

In view of the foregoing a simple explanation is obtained for the negative temperature coefficient

of resistivity in the interval 310—330°K which includes the transition temperature T_S. Since the transition does not occur entirely at a single point T_S but is spread over an interval of about 20°, it is reasonable to presume that the semiconducting character of conductivity in the region of T_S results from a heterophase state rather than from a gap in the electron energy spectrum at the antiferromagnetic-ferromagnetic transition. As the temperature is increased in the region of T_S the resistivity decreases because of the increasing amount of matter in the ferromagnetic phase, which exhibits considerably higher electric conductivity than the antiferromagnetic phase.

The different character of the temperature and field dependences of galvanomagnetic effects in ferromagnetic and antiferromagnetic regions having the same crystalline structure indicates that the character of spin ordering is of essential importance for these effects.

It should be noted in conclusion that similar electric and galvanomagnetic properties, and their anomalies, are observed in a large group of rare-earth metals undergoing a magnetic transition at Θ_2 from the helicoidal antiferromagnetic state to the paramagnetic state.[10]

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