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THE EFFECT OF PRESSURE ON THE ELECTRICAL RESISTANCE AND GALVANOMAGNETIC EFFECT IN CHROMIUM TELLURIDE

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The temperature dependences of the electrical resistance and of the galvanomagnetic effect were measured in chromium telluride near the magnetic transition temperature at a pressure of 4,600 kg/cm^2. The shift in the Curie point under hydrostatic compression of the specimen was determined [Eq. (1)]. The variation in the exchange integral with interatomic distance in the Cr–Te system was also studied by measuring the electrical, magnetic, and galvanomagnetic properties of solid solutions of Cr–Te–Se. On the basis of the data obtained it is deduced from the thermodynamics of ferromagnetism that the nature of the change in spontaneous magnetization of CrTe is significantly different, depending on whether the reduction in the unit cell volume is produced by the hydrostatic pressure or by the introduction of selenium impurity.

INTRODUCTION

THE transition metals chromium and manganese form compounds with the elements of the subgroups V and VI-B which have the same crystal structure but different magnetic properties: ferromagnetic (CrTe, MnSb), antiferromagnetic (CrSb, CrSe, MnTe) or ferrimagnetic ($CrS_{1.17}$). There have recently been several experimental[1-3] and theoretical[4] studies of an interesting property of these compounds, which is the change in the spin ordering on substituting one of the components of these binary alloys by another of the same system, giving rise to solid solutions of the type $CrTe_{1-x}Se_x$, $CrTe_{1-x}Sb_x$, $MnCr_{1-x}Sb_x$, and $Mn_{1-x}Cr_xTe$. The change in the relative concentrations of the components of a ternary alloy leads to a change in spin ordering from ferromagnetic to antiferromagnetic (or the reverse), accompanied by a reduction in the magnetic transition temperature and a change in the lattice parameters. The transition from the ferromagnetic to antiferromagnetic state in these systems is related to the reduction in unit cell volume of the crystal.

In the present work an attempt is made to separate the influence of two factors on the spin ordering: volume changes and the change in the nature of the atomic neighbors. In order to study the change in Curie temperature Θ_f and in the spontaneous magnetization σ_s of chromium telluride under the influence of pressure, the temperature dependence of electrical resistance and the isotherms of the galvanomagnetic effect $\Delta R/R$ were measured in the region of the magnetic transition temperature at a pressure of 4,600 kg/cm^2 (we shall use $r = \Delta R/R$ for brevity). At the same time measurements on the ternary alloy $CrTe_{1-x}Se_x$ were carried out. The changes in Θ_f, σ_s, r, the resistivity ρ, and the elementary cell volume were determined as functions of selenium concentration ($0 \leq x \leq 0.1$), and the temperature dependences of σ_s, r and ρ of these alloys were investigated in the neighborhood of the Curie point.

APPARATUS AND METHOD OF MEASUREMENT

The high-pressure vessel used in the present work was made of 60G8N8Kh3V high-strength austenitic steel. A preliminary test of the vessel, by producing a hydrostatic pressure of up to 5,200 kg/cm^2 and then measuring the strength of the magnetic field inside its channel, showed that the deformation produced by this pressure in the cold state does not lead to the appearance of the ferromagnetic martensitic phase.

The specimen was contained in a 3.9 mm diameter channel in the lower part of the chamber, which in turn was 36 mm in diameter. This part of the chamber was placed between the poles of an electromagnet with a gap of 41 mm. Five conical type electrical leads were mounted on the upper part of the chamber on a special seal with copper packing rings. We used one lead for measuring the electrical resistance of a manganin manometer, in the form of a bifilar loop, laid in a groove of a special holder to which the specimen was fastened. The body of the chamber was used as the second lead for the manganin manometer.

To avoid additional electrical connections for measuring the temperature inside the chamber, the remaining four leads were used both for measuring the electrical resistance of the specimen and for temperature measurement. For this purpose, two copper-constantan thermocouples were attached to the ends of the specimen, the copper arms acting as current leads and the constantan as potential probes for the specimen. The cold junctions of the thermocouples were at atmospheric pressure and 0° C. By this simple arrangement the number of leads could be restricted to four, instead of the eight necessary for this sort of measurement.

The thermal emf of the thermocouples and the potential drop along the specimen were measured with a type PPTN-1 low resistance potentiometer and a M-21/IV mirror galvanometer with sensitivity 10^{-7}v/mm. While the thermocouple emf was being measured the primary current through the specimen was turned off so that there should be no Peltier effect at the junction of the specimen with the copper leads.

We did not take account of the effect of pressure on the thermocouple emf's since a control measurement of the emf of a "pressure thermocouple," made with arms of compressed and uncompressed metals, showed that the error introduced into the temperature determination can be neglected, for it does not exceed 0.024°. To remove errors due to extraneous effects, the measurements were made with the compensated current flowing in both directions, and the change of resistance in the magnetic field was determined for both directions of the field and of the current in the specimen. The maximum field strength was 8000 oe.

The hydrostatic pressure in the chamber was produced by a high pressure compressor using the system of L. F. Vereshchagin. Transformer oil was used as pressure transmitter. For thermostating, the chamber was surrounded by a brass case, through which was passed water from a Geppler ultra-thermostatic bath. In order to check on the absence of irreversible changes produced in the specimen by the pressure, control measurements of the temperature dependence of $\Delta R_{\perp}/R$ at atmospheric pressure and a field of 8000 oe were made at the end of a cycle of high pressure measurements.

EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows the results of the measurements of the effect of hydrostatic pressure on the electrical resistivity of chromium telluride at room temperature; points corresponding to increasing and decreasing pressure are shown by different symbols. It can be seen from the graph that the measurements with pressure increasing and decreasing give practically identical results: the resistivity increases with pressure, with the pressure coefficient $R_T^{-1}dR/dp$ equal to (1 — 1.5) $\times 10^{-4}$ $kg^{-1}cm^2$ within the range of measurement, which is two to three orders of magnitude greater than known values of $R_T^{-1}dR/dp$ for ferromagnetic metals and alloys.[5,6]

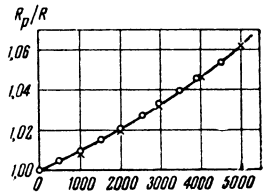

FIG. 1. The influence of pressure on the electrical resistivity of CrTe at room temperature; × – pressure increasing, ○ – pressure decreasing.

From the sign and magnitude of the pressure coefficient of resistivity it could be expected that hydrostatic compression should shift the Curie point of chromium telluride to lower temperatures, and that the magnitude of this effect should be fairly great. On this basis we tried to determine $d\Theta_f/dp$ from direct measurements of the temperature dependence of the electrical resistivity at high pressure, and not to calculate the effect from the measured temperature and pressure coefficients of resistivity in the region of the Curie point, as is usually done for small changes of Θ_f with pressure.[6]

Figure 2 shows the results of measuring the temperature dependence of resistivity at atmos-

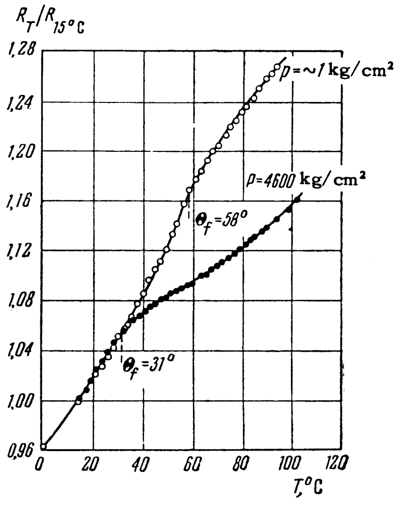

FIG. 2. The temperature dependence of the electrical resistivity of CrTe at atmospheric pressure and at p = 4,600 kg/cm².

pheric pressure and at 4600 kg/cm^2. It can be seen from the curves that the temperature of the Curie point, determined from the break in the R(T) curve, is +58° C at atmospheric pressure and +31° C at 4600 kg/cm^2. Repeated measurements showed good reproducibility of these data, giving the change in Curie point of chromium telluride under hydrostatic pressure as:

$$d\Theta_f/dp = (-5.9 \pm 0.3)\cdot 10^{-3}\ \text{deg-kg}^{-1}\text{cm}^2. \qquad (1)$$

The value of $d\Theta_f/dp$ was checked by measuring the galvanomagnetic effect $r = \Delta R/R$ at high pressures. The change in resistivity of CrTe in a magnetic field at $p = 4600$ kg/cm^2 was determined both above and below the Curie point. The isotherms of the galvanomagnetic effect r(H) obtained from these measurements are shown in Fig. 3. Figure 4 shows the curves of $r_\perp(t)$, representing the temperature dependence of the transverse galvanomagnetic effect at atmospheric pressure and at 4,600 kg/cm^2 in a magnetic field of 8000 oe. The change in Curie temperature under the influence of the hydrostatic pressure was determined in this case from the shift in the maximum of the galvanomagnetic effect $(-r)_{\max}$ with the result, $d\Theta_f/dp = -6.2 \times 10^{-3}$ deg-kg^{-1}cm^2.

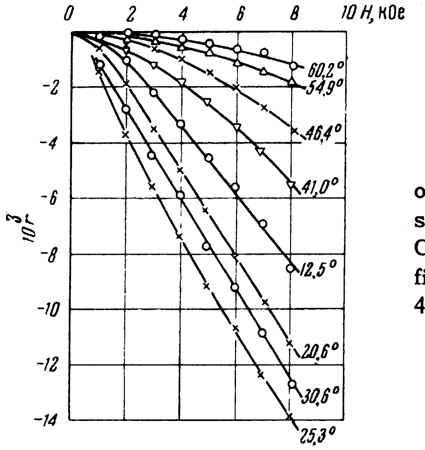

FIG. 3. Isotherms of the electrical resistivity change of CrTe with magnetic field at a pressure of 4,600 kg/cm^2.

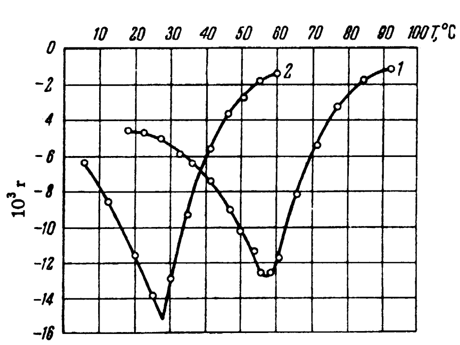

FIG. 4. Temperature dependence of the galvanomagnetic effect: curve 1 – at atmospheric pressure, curve 2 – at a pressure of 4,600 kg/cm^2.

We should point out that the value of $d\Theta_f/dp$ obtained is close to the magnitude of the effect in the invar alloys[7] (30% Ni, 70% Fe) and is the largest among all substances so far investigated.

Using the value of the compressibility* of chromium telluride $\kappa = (22 \pm 3) \times 10^{-7}$ cm^2/kg we found $d\Theta_f/dV = 3.2 \times 10^{25}$ deg-cm^{-3}.

It seemed of interest to compare the data obtained with the results of other measurements, in which the reduction in elementary cell volume is produced not by hydrostatic pressure but by introducing impurities with formation of substitutional solid solutions. The change in the Curie temperature of CrTe connected with the reduction in the interatomic distance on substituting tellurium atoms by selenium ($\text{CrTe}_{1-x}\text{Se}_x$) has been studied earlier by Tsubokawa[1] and by Lotgering and Gorter.[3] The results, while agreeing qualitatively with one another and with our data, disagree violently with one another quantitatively. For example, the values of Θ_f for chromium telluride given by different authors differ by 30° from one another and those of $d\Theta_f/dV$ differ by 37 – 40%.

We used three different methods for determining Θ_f in order to obtain more reliable data on the magnetic transition temperature of $\text{CrTe}_{1-x}\text{Se}_x$ alloys and on its variations with the elementary cell volume of the crystal: (1) from the break in the resistivity-temperature curve, (2) from the maximum in the galvanomagnetic effect $r_{\max}$, and (3) from the disappearance of spontaneous magnetization, determined by the method of "thermodynamic coefficients" ($T = \Theta_f$ for $\alpha = 0$).[9]

The elementary cell volume of the alloys was determined by x-ray diffraction. The diffraction patterns were taken on a KROS-1 camera using a BSVL electron tube with chromium anticathode, with aluminum as the reference. The lattice parameters were calculated from the interference lines $(203)\alpha_1$, $(211)\alpha_1$ of the specimen and the lines $(113)\beta_1$ and $(222)\beta_1$ of the standard. These parameters and the elementary cell volumes of the $\text{CrTe}_{1-x}\text{Se}_x$ alloys are shown in Table I, together with the Curie temperatures determined by the three methods indicated above.

From the data of Table I, $d\Theta_f/dV$ was calculated as 3.5×10^{25} deg-cm^{-3}, which agrees (within the limits of error of measuring the compressibility) with the value obtained in the pressure experiments. From this we can conclude that the interaction exchange integral in the Cr-Te

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*The compressibility of CrTe was measured by Yu. A. Bazhin by a tensometric method.[8] We take this opportunity to thank him.

Table I. The lattice parameters a and c, the unit cell volume V and the Curie temperature Θ_f for $CrTe_{1-x}Se_x$ alloys

Composition	a, kX	c, kX	c/a	V, kX³	Θ_f, °C		
					1	2	3
CrTe	3.985	6.268	1.573	86.20	58.0	57.5	60.0
$CrTe_{0,93}\ Se_{0,07}$	3.974	6.241	1.570	85.35	29.5	27.0	35.0
$CrTe_{0,90}\ Se_{0,10}$	3.963	6.233	1.573	84.77	20.5	19.0	22.0

system changes proportionally with the reduction in elementary cell volume, independently of whether this reduction is produced by hydrostatic pressure or by the introduction of selenium impurity with the formation of solid substitutional solutions $CrTe_{1-x}Se_x$.

In order to decide whether hydrostatic pressure produces, besides a reduction in the interaction exchange integral, a change in the magnetic moment of the chromium atom in CrTe, we shall consider in more detail the isotherms of the galvanomagnetic effect which we measured at a pressure of 4,600 kg/cm². According to our measurements, the sign of the effect is negative, regardless of the relative direction of the current in the specimen and of the external field ($r < 0$ both for $H \parallel I$ and for $H \perp I$), which can be explained by the influence of the paraprocess, since the r(H) curves obtained experimentally by us agree with the theoretical relations proposed by Akulov:[10]

$$r = aH^{2/3} \quad \text{for} \quad T = \Theta_f,$$
$$r = bH^2 \quad \text{for} \quad T > \Theta_f. \tag{2}$$

The law relating the galvanomagnetic effect to the magnetic field strength at the Curie point, analogous to the first equation of (2), can be obtained on the basis of the thermodynamic theory of ferromagnetism,[12] as was shown by Belov and Zaĭtseva,[11] using the relation found by Gerlach[13]

$$r_s = c\sigma_s^2, \tag{3}$$

where r_s is the value of the galvanomagnetic effect determined by the spontaneous magnetization. The coefficient a is then*

$$a = c\beta^{-2/3}\sigma_0^{8/3}. \tag{4}$$

Here β is a thermodynamic coefficient depending on pressure and temperature and σ_0 is the absolute saturation magnetization.

Figure 5 shows the variation of r with $H^{2/3}$ for chromium telluride at atmospheric pressure and at a pressure of 4,600 kg/cm², and also for the alloy $CrTe_{0.93}Se_{0.7}$, the elementary cell volume of which is equal to the elementary cell volume of CrTe at a pressure of 4,600 kg/cm². Because of the strong dependence of the galvanomagnetic effect on temperature, the values of r are given for the same values of reduced temperature $T/\Theta_f = 0.99$. It can be seen that there is in all cases a linear dependence of r on $H^{2/3}$.

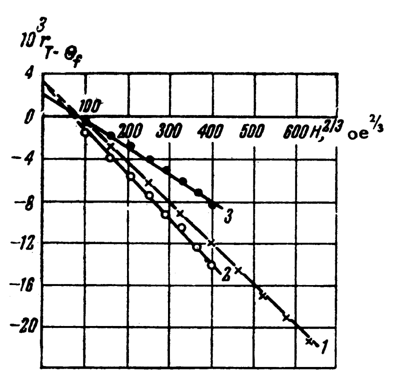

FIG. 5. The dependence of r on $H^{2/3}$: curve 1 – CrTe at atmospheric pressure, curve 2 – CrTe at p = 4,600 kg/cm², curve 3 – $CrTe_{0.93}Se_{0.07}$. at atmospheric pressure.

The values of the coefficient a which characterize the slope of the straight lines and determine the intensity of the paraprocess, and also the magnitudes of the spontaneous galvanomagnetic effect r_s, corresponding to the positive intercepts on the ordinate axis, are shown in Table II. This also gives our measured values of spontaneous magnetization σ_s and of resistivity ρ. We took for σ_0 the experimental values[3] of the saturation magnetization at T = 20° K. We determined the temperature dependence of σ_s of the alloy $CrTe_{0.93}Se_{0.07}$ in the region of the Curie point by two methods: from the $r(\sigma^2)$ curves, extrapolation of the linear parts of which to the value $r = 0$ gives the square of the spontaneous magnetization, and also by the method of "thermodynamic coefficients,"[9] which

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*Equation (4) for the coefficient a differs from the analogous expression for a in Eq. (7) of Belov and Zaĭtseva's paper[11] in that in its derivation a term depending on the external magnetic field as σH was included in the expansion of the thermodynamic potential as a power series in the relative magnetization. In references 9 and 11 this term was included in the form ηH, which does not agree with the dimensions of thermodynamic potential, but corresponds to the energy of the magnetic field leading to saturation magnetization.

Table II

Composition	$10^5 a$	$10^3 r_s$	σ_s Gauss-cm^3/g	σ_0 Gauss-cm^3/g	$10^{-4}\rho$, ohm-cm
			$T/\Theta_f = 0.99$	$T = 20°K$	$T = 291°K$
CrTe($p = \sim 1$ kg/cm^2)	3.8	3.3	14.2	76.3	2.1
CrTe($p = 4600$ kg/cm^2)	4.3	3.3	—	—	3.5
$CrTe_{0.93}Se_{0.07}$	2.6	2.3	10.1	75.0	8.2

made possible the simultaneous determination of the temperature dependence of the thermodynamic coefficient β.

Comparison of the results of measuring $CrTe_{0.93}Se_{0.07}$ with the earlier study of the temperature variation of spontaneous magnetization in CrTe[14] indicates that the magnetic transition in $CrTe_{0.93}Se_{0.7}$ is more spread out. The coefficient ξ in the well known relation $(\sigma_s/\sigma_0)^2 = \xi(1 - T/\Theta_f)$ is 1.44 for $CrTe_{0.93}Se_{0.07}$, which is considerably smaller than the value of ξ obtained for CrTe.

The dependence of the galvanomagnetic effect r on the reduced temperature T/Θ_f for CrTe at atmospheric pressure and at 4,600 kg/cm^2 and also for the alloy $CrTe_{0.93}Se_{0.07}$ is shown in Fig. 6. In all three cases the values of r are taken from the isotherms of the galvanomagnetic effect at a field strength H = 8000 oe. It follows that for $T < \Theta_f$, pressure leads to an increase in the absolute magnitude of the effect in CrTe, while for $T > \Theta_f$ (in the paramagnetic region) the $r(T/\Theta_f)$ curves at atmospheric pressure and at p = 4,600 kg/cm^2 practically coincide. This indicates that the observed change in the galvanomagnetic effect under pressure is connected with a change in magnetization produced by hydrostatic compression. The curve obtained for $CrTe_{0.93}Se_{0.07}$ lies appreciably below the first two over the whole temperature range examined.

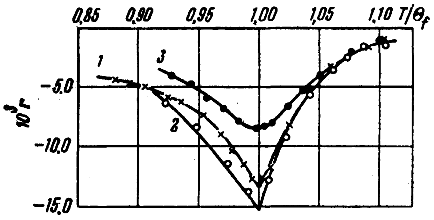

FIG. 6. Temperature dependence of the transverse galvanomagnetic effect at H = 8000 oe: curve 1 – CrTe at atmospheric pressure, curve 2 – CrTe at p = 4,600 kg/cm^2, curve 3 – $CrTe_{0.93}Se_{0.07}$ at atmospheric pressure.

It can be seen from the data given in Table II and Figs. 5 and 6 that the nature of the change in the galvanomagnetic effect is, to a considerable extent, dependent on whether the hydrostatic compression of the specimen is brought about by the action of pressure or by the formation of $CrTe_{1-x}Se_x$ solid solutions. The difference is seen primarily in the considerable increase in the intensity of the paraprocess produced by pressure, while the magnitude of the spontaneous galvanomagnetic effect r_s remains unchanged. Reduction of the elementary cell volume by introduction of selenium impurity leads to a reduction both in the intensity of the paraprocess, in r_s and in σ_0. The value of the electrical resistivity then increases fourfold.

If we assume that the coefficient c, which determines the relation between r and the square of the spontaneous magnetization [Eq. (3)], does not change with pressure*, then we can conclude that the whole change in spontaneous magnetization in CrTe produced by hydrostatic pressure is brought about only by a change in the exchange integral, with a constant value of the absolute saturation magnetic moment. According to (4) we can say that the observed increase in the intensity of the paraprocess under pressure is related to a decrease in the thermodynamic coefficient β with σ_0 constant. Kondorskii and Sedov[15] considered the reasons for a reduction in the absolute saturation magnetization in ferro- and ferrimagnetic substances under the influence of pressure. If we assume, according to earlier deductions,[14] that CrTe is a ferromagnetic with small s-d exchange interaction, then we may consider that the pressures employed do not bring about electron transitions from the s to the d band.

The decrease in the intensity of the paraprocess in $CrTe_{0.93}Se_{0.07}$ in contrast with CrTe (at p = 4,600 kg/cm^2) is related both to a decrease in the absolute saturation magnetization σ_0 and to an increase in the thermodynamic coefficient β. We can explain the appreciable increase in ρ in the following way. Realizing that CrSe is antiferromagnetic, we can assume that substitution of tellurium atoms by selenium in the CrTe com-

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*To some extent we can consider the basis for such an assumption to be the fact that there is only a small difference in the coefficient c (up to 2%) as determined by us for CrTe and for $CrTe_{0.93}Se_{0.07}$ from the $r(\sigma^2)$ curves for the same values of T/Θ_f.

pound will lead to the existence of chromium ions with antiparallel spins; the indirect nature of the exchange interaction with the active part of the Se atoms must be dominant in this. The random distribution of ions with antiparallel spins leads to an increase in electrical resistivity of the alloy $CrTe_{0.93}Se_{0.07}$ due to scattering of conduction electrons by the fluctuations of magnetic moment.

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