

## MEASUREMENTS OF THE MAGNETIZATION OF $\text{CrTe}_{1-x}\text{Sb}_x$ ALLOYS IN STRONG PULSED MAGNETIC FIELDS

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Magnetization isotherms of  $\text{CrTe}_{1-x}\text{Sb}_x$  alloys with different Sb contents ( $0 \leq x \leq 0.9$ ) were measured. The measurements were carried out in pulsed magnetic fields of intensity up to 300 kOe in the temperature interval from 77 to 380°K. The magnetic phase diagram constructed on the basis of the results does not agree in the concentration region  $0 \leq x \leq 0.5$  with the results of neutron-diffraction investigations<sup>[2]</sup> and with the de Gennes theory<sup>[1]</sup> based on the double-exchange mechanism. Alloys with larger Sb contents ( $0.6 < x < 0.9$ ) have at 77°K a spontaneous moment and a nonzero differential susceptibility in strong fields. Qualitative estimates of the values of the transport integral and of the ratio of the width of the band to the exchange-interaction energy of these alloys are given.

### 1. INTRODUCTION

THE magnetic state of the solid solutions  $\text{CrTe}_{1-x}\text{Sb}_x$  changes with increasing Sb content, i.e., with increasing  $x$ , from ferromagnetic (CrTe) to antiferromagnetic (CrSb). This system of alloys is an interesting research object for verifying the de Gennes theory of double exchange and for clarifying the role of current carriers in the stabilization of any magnetic structure.

According to the de Gennes theory, the double exchange due to the presence of current carriers leads to a distortion of the spin configuration of the ground state, to the occurrence of a canted magnetic structure, and, as a consequence, also to specific magnetic properties not observed in ordinary ferromagnetic and antiferromagnetic substances. These peculiarities are manifest in the occurrence of a small spontaneous moment at low temperatures, in the absence of saturation in strong magnetic fields, and in the existence of several magnetic-transformation temperatures in the same alloy. Namely, besides transitions of the type of the Curie point ( $\theta_f$ ) and of the Neel point ( $\theta_N$ ), a lowering of the temperature should lead also to magnetic transformations (at a temperature ( $T_g$ ) connected with the transition of ferro- or antiferromagnetic ordering into canted structures.

An experimental verification of the indicated conclusions of the theory revealed certain contradictions. Thus, neutron-diffraction investigations<sup>[2]</sup> of  $\text{CrTe}_{1-x}\text{Sb}_x$  solid solutions and the establishment of a change in the magnetic structure of these systems were in full agreement with the de Gennes model and with the generalized magnetic phase diagram established by him. However, an investigation of the electric properties of the alloys<sup>[3]</sup>, carried out by one of the present authors, has contradicted the conclusions of the theory and has cast doubt on the possibility of using the de Gennes model for the indicated system. In addition, there was ambiguity also in the determination of the magnetic structure of chromium telluride and of the compounds  $\text{Cr}_5\text{Te}_6$  and  $\text{Cr}_3\text{Te}_4$ , which are close to it in chemical composition. The (001) reflection observed in the neu-

tron diffraction patterns at low temperatures could be attributed with equal probability to an orthorhombic distortion of the crystal structure<sup>[4]</sup> or to a canted magnetic structure<sup>[2]</sup>.

The need therefore arises for an additional and independent experimental verification of the magnetic diagram of state and of the magnetic structures of the indicated alloys. It is possible to use for this purpose measurements of the magnetization in strong pulsed magnetic fields, the results of which are reported in the present article.

### 2. MEASUREMENT PROCEDURE AND RESULTS

The  $\text{CrTe}_{1-x}\text{Sb}_x$  solid solutions were synthesized by heating thoroughly mixed powders of the initial chemically-pure elements Cr, Te, and Sb in evacuated quartz ampoules. The thermal conditions of the synthesis were analogous to those described in<sup>[5]</sup>. Control measurements of the specific magnetization, of the Curie points, and also an x-ray diffraction determination of the crystal unit-cell parameters agreed with the data published in<sup>[5]</sup>.

Particular attention was paid to the phase composition of the  $\text{CrTe}_{1-x}\text{Sb}_x$  alloys, to which end x-ray diffraction and microstructure analyses were performed. The analysis data has shown that the investigated samples were single-phase.

The magnetic measurements in pulsed magnetic fields were carried out by the Faraday method, using the piezoelectric pulse balance described in<sup>[6]</sup>. The time required for the magnetic field to rise to the first maximum value was 75  $\mu\text{sec}$ . Special measures were adopted to eliminate the measurement errors due to the occurrence of eddy currents in the sample. To this end, the initial  $\text{CrTe}_{1-x}\text{Sb}_x$  alloys were ground into powder and mixed with BF-2 resin in such proportions that after drying the sample had an increased resistance not less than  $10^6$  ohm. The pulsed magnetic balance was calibrated for each investigated sample by comparing the relative values of the magnetization  $\sigma$ , obtained by measurements in the pulsed regime, with the values of

$\sigma$  obtained in static measurements at the same temperatures. The error in the measurement of the magnetization did not exceed 3%.

The Curie temperature and the spontaneous magnetization in the region of the magnetic transformation were determined by the method of thermodynamic coefficients developed by Belov<sup>[7]</sup>. In this case the magnetization isotherms were measured in static fields up to 19 kOe with the aid of a Domenicali-type magnetometer<sup>[8]</sup>. The obtained data were processed in the form of plots of  $H/\sigma$  against  $\sigma^2$ , so as to be able to compare the well known thermodynamic equation (see<sup>[9]</sup>)

$$\alpha\sigma + \beta\sigma^3 = H \quad (1)$$

with the results of our measurements.

We measured 9  $\text{CrTe}_{1-x}\text{Sb}_x$  samples with different Sb contents ( $x = 0, 0.1, 0.2, 0.5, 0.6, 0.7, 0.8, \text{ and } 0.9$ ).

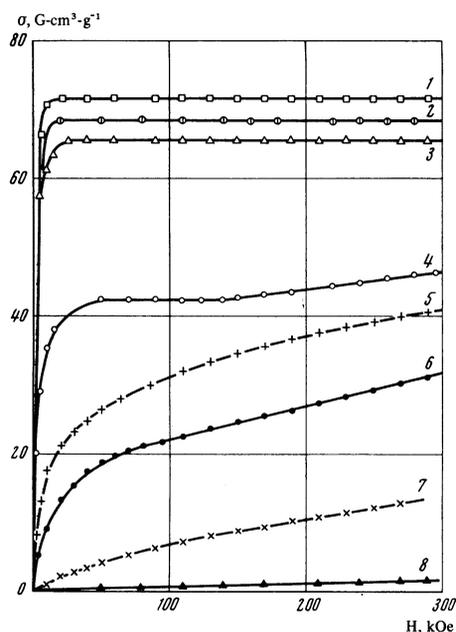


FIG. 1. Magnetization isotherms of  $\text{CrTe}_{1-x}\text{Sb}_x$  alloys with different Sb concentrations at  $T = 77^\circ\text{K}$ : 1-0, 2-0.1, 3-0.2, 4-0.5, 5-0.6, 6-0.7, 7-0.8, 8-0.9.

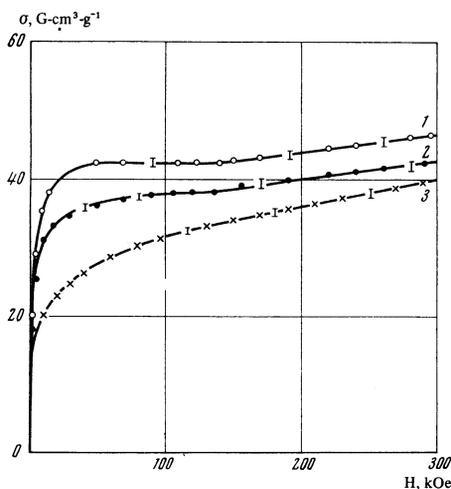


FIG. 2. Magnetization curve of the alloy  $\text{CrTe}_{0.5}\text{Sb}_{0.5}$  at temperatures ( $^\circ\text{K}$ ): 1-79, 2-128, 3-178.

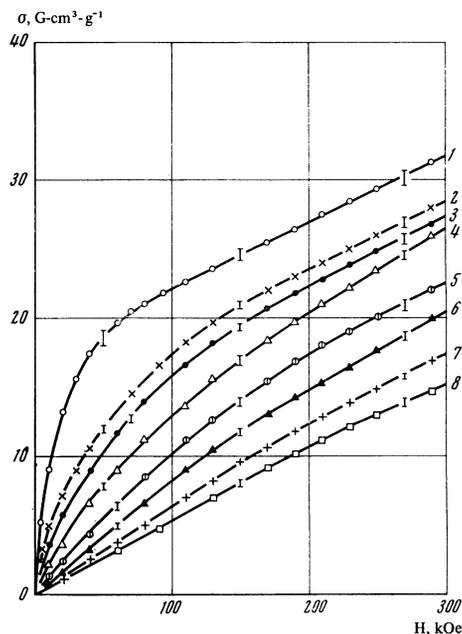


FIG. 3. Magnetization isotherms of the alloy  $\text{CrTe}_{0.3}\text{Sb}_{0.7}$  at temperatures ( $^\circ\text{K}$ ): 1-77, 2-132, 3-143, 4-165, 5-191, 6-213, 7-233, 8-268.

Figure 1 shows the magnetization isotherms of these alloys, measured at liquid-nitrogen temperature in fields up to 300 kOe. It is seen from the result that with increasing Sb concentration, i.e., with increasing  $x$ , the magnetization  $\sigma$  decreases, the change being particularly strong at large  $x$ .

Based on the character of the  $\sigma(H)$  plots in Fig. 1, all the measurement alloys can be divided into four groups.

The first is  $\text{CrTe}$  and compositions close to it with  $x = 0.1$  and  $x = 0.2$ . It is characterized by saturation of the magnetization in fields above 20 kOe (curves 1, 2, and 3).

The second group comprises the alloy  $\text{CrTe}_{0.5}\text{Sb}_{0.5}$  (curve 4, Fig. 1), for which saturation occurs in stronger fields (40-50 kOe) and for which a particularly remarkable fact is the linear increase of the specific magnetization when a certain field value  $H_c$ , approximately equal to 150 kOe, is reached. The differential susceptibility above this field, calculated by least squares, is  $2.0 \times 10^{-5}$  and is independent of the temperature within the limits of the experimental accuracy (curves 1 and 2 of Fig. 2).

The third group includes the alloys with large Sb contents:  $x = 0.6, 0.7, \text{ and } 0.8$  (curves 5, 6, and 7 of Fig. 1). In this case saturation of the magnetization is not reached in the entire investigated field interval, and the differential susceptibility is quite large,  $\chi_{\text{dif}} = (3.6-4.8) \times 10^{-5}$ , and depends on the temperature of the experiment. This is seen from the curves of Fig. 3, which represent the magnetization isotherms of  $\text{CrTe}_{0.3}\text{Sb}_{0.7}$ , plotted in the temperature interval from 77 to 268°K. Extrapolation of the resultant curves from 150-300 kOe to a zero field makes it possible to determine the spontaneous magnetization  $\sigma_s$ , which turns out to be much smaller than the nominal value obtained for  $\text{CrTe}$  and calculated from the Brillouin function at  $j = 3/2$  (see

Fig. 6 below). It is important to note the smooth increase of  $\sigma_s$  with increasing temperature, indicating that the magnetic phase transition is a diffused one.

The last, fourth group includes alloys close in their chemical composition to the antiferromagnetic compound CrSb. Figure 1 (curve a) shows the dependence of the magnetization on the magnetic field intensity for the alloy CrTe<sub>0.1</sub>Sb<sub>0.9</sub>, from which it is seen that  $\sigma$  increases linearly with H and that the susceptibility is equal in this case to  $\chi = 6 \times 10^{-6}$ . Measurements performed at higher temperatures (153–383°K) have shown that the value of  $\chi$  of this alloy changes little with T.

Thus, the features of the variation of magnetization in strong pulsed magnetic fields make it possible to subdivide the entire system of CrTe<sub>1-x</sub>Sb<sub>x</sub> solid solutions into four groups which, as will be shown below, differ from one another also in the character of the magnetic transformation in the region of the Curie point. To this end, we investigated the temperature dependence of the spontaneous magnetization, and, using the equation of the magnetic state

$$(\sigma_s / \sigma_0)^2 = \xi(1 - T / \Theta_f), \quad (2)$$

we determined the rate of the magnetic transformation with the aid of the coefficient  $\xi$ . These data, alongside other magnetic characteristics of the CrTe<sub>1-x</sub>Sb<sub>x</sub> alloys, are given in Table I and in Fig. 4, which shows the temperature dependence of the square of the relative spontaneous magnetization for three different alloys with  $x = 0, 0.2, \text{ and } 0.5$ . From the presented data we see that the alloys of the first and second groups differ from one another in the rate of magnetic transformation. According to the theory of Vonsovskii and Vlasov<sup>[10]</sup>, for metallic ferromagnets we have  $\xi > 3$ , while for ferrites this coefficient is much smaller than 3 and, as shown by experiment<sup>[7]</sup>, lies in the range

Table I. Magnetic characteristics of CrTe<sub>1-x</sub>Sb<sub>x</sub> alloys

Alloy	$\Theta_f, ^\circ\text{K}$	$\Theta_p, ^\circ\text{K}$	$M_f, \mu_B$	$M_p, \mu_B$	g-factor	$\xi$
CrTe	342	344	2.34	4.9	2.01	2.46
CrTe <sub>0.9</sub> Sb <sub>0.1</sub>	319	320	2.31	4.7	2.005	2.20
CrTe <sub>0.8</sub> Sb <sub>0.2</sub>	299	300	2.25	4.6	2.005	2.04
CrTe <sub>0.5</sub> Sb <sub>0.5</sub>	249	234	1.42	4.4	2.00	0.70
CrTe <sub>0.4</sub> Sb <sub>0.6</sub>	249	233	—	4.2	—	0.71

Note.  $\Theta_f$ —Curie point;  $\Theta_p$ —paramagnetic Curie temperature;  $M_f$  and  $M_p$ —values of the effective magnetic moments calculated from the saturation magnetization and in accordance with the Curie-Weiss law; measurements of the g-factor by the EPR method were made on our samples by V. K. Kapustkin;  $\xi$ —coefficients determining the rate of magnetic transformation.

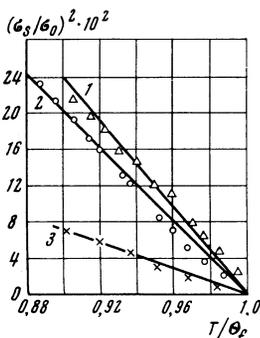


FIG. 4. Temperature dependence of the square of the relative spontaneous magnetization of CrTe<sub>1-x</sub>Sb<sub>x</sub> alloys with different Sb concentrations near the Curie temperature: 1— $x = 0$ , 2—0.2, 3—0.5.

0.1–0.7. The value  $\xi = 2.46$  obtained by us for CrTe corresponds to the theoretical value calculated from the formulas of the quasiclassical theory of ferromagnetism, in which no account is taken of the spontaneous magnetization of the conduction electrons. The value  $\xi = 0.7$  of the alloy CrTe<sub>0.5</sub>Sb<sub>0.5</sub> is the same as in most ferrites.

The alloys of the third group (in the concentration region  $0.6 \leq x \leq 0.9$ ) have two magnetic transformation points,  $T_S$  and  $\Theta_N$ . It has been established that the low-temperature magnetic transformation point  $T_S$  is connected with the gradual vanishing of the spontaneous magnetization, but the magnetic behavior of the alloys in this temperature region has not been investigated in detail, so that the nature of this transformation is not yet clear. This may be a transformation connected with the spin disordering of the ferromagnetic phase, or else a transformation of the canted magnetic structure into an antiferromagnetic one, as predicted by the de Gennes theory. The high-temperature transformation point  $\Theta_N$  apparently corresponds to an antiferromagnetism—paramagnetism transition, since the anomalies of the temperature dependence of the susceptibility in this region are very similar to the behavior of antiferromagnets at the Neel point<sup>[5]</sup>.

We investigated three compositions of this group of alloys ( $x = 0.6, 0.7, \text{ and } 0.8$ ), paying special attention to the character of the magnetic behavior in the region of the low-temperature transformation. We have established the following: at liquid-nitrogen temperature, all the indicated alloys have a small spontaneous moment, which decreases slowly with increasing temperature. For CrTe<sub>0.4</sub>Sb<sub>0.6</sub>, the character of the magnetic transformation, its rate, and the transition temperature itself are exactly the same as for CrTe<sub>0.5</sub>Sb<sub>0.5</sub>, indicating the presence of a single ferromagnetic phase in this solid-solution region.

The low-temperature magnetic transformation of alloys with large Sb concentration ( $x = 0.7, \text{ and } x = 0.8$ ) differs in character from all the alloys considered above. This difference is expressed in the fact that in the region of the magnetic transition, the change of the magnetization with changing field does not follow the thermodynamic equation (1), which describes the transformation of ferro- and antiferromagnets at the Curie point. This is clearly seen from the curves in Fig. 5,

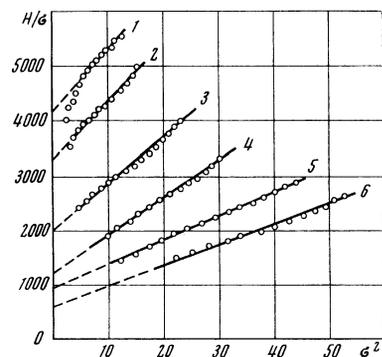


FIG. 5. Dependence of  $H/\sigma$  on  $\sigma^2$  for CrTe<sub>0.3</sub>Sb<sub>0.7</sub> at the following temperatures (°K): 1—173.2; 2—165.3, 3—154.4, 4—143.4, 5—132.3, 6—122.7.

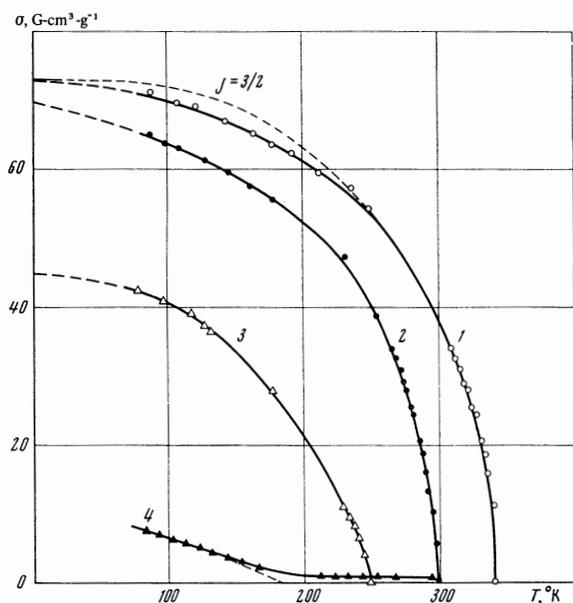


FIG. 6. Temperature dependence of the spontaneous magnetization for alloys with different compositions: 1—CrTe, 2—CrTe<sub>0.8</sub>Sb<sub>0.2</sub>, 3—CrTe<sub>0.5</sub>Sb<sub>0.5</sub>, 4—temperature dependence of the specific magnetization of CrTe<sub>0.3</sub>Sb<sub>0.7</sub> in a field of 8 kOe. Dashed line—Brillouin function for  $j = 3/2$ .

which represent the magnetization isotherms of CrTe<sub>0.3</sub>Sb<sub>0.7</sub> in the form of  $H/\sigma = f(\sigma^2)$  plots. Attention is called to the fact that the condition for the Curie point, whereby the thermodynamic coefficient  $\alpha$  should be negative in the entire region of existence of spontaneous magnetization and zero at  $T = \theta_f$ , is not satisfied in this case. It is seen from the plots of Fig. 5 that  $\alpha$  has a positive sign in the temperature interval from 122.7 to 173.2°K, although, judging from the  $\sigma = f(T)$  curve measured at  $H = 8$  kOe (Fig. 6, curve 4), the magnetic transformation occurs near 180°K, and in this temperature region one could expect a change in the sign of the coefficient  $\alpha$ . These data indicate that the low-temperature magnetic transition in alloys with large Sb concentration ( $x = 0.7$  and  $x = 0.8$ ) is not a magnetic transformation of the Curie-point type, and is apparently connected with a change of the magnetic structure.

### 3. MAGNETIC DIAGRAM OF STATE AND DISCUSSION OF THE RESULTS

On the basis of the results, let us explain the general course of the magnetic phase diagram of the CrTe<sub>1-x</sub>Sb<sub>x</sub> solid solutions. First to be noticed is the disparity between our experimental data in the concentration interval  $0 \leq x \leq 0.5$  and the predictions of the theory<sup>[1]</sup> or the neutron-diffraction results<sup>[2]</sup>. According to the cited works, one should expect at low temperature a nonzero differential susceptibility in strong fields for the entire system of CrTe<sub>1-x</sub>Sb<sub>x</sub> solid solutions<sup>1)</sup>. Our experi-

mental data indicate, however, that in CrTe and in the compositions adjacent to it there is magnetization saturation, and only in the alloys with large concentrations of Sb ( $x \geq 0.6$ ) is a sufficiently large differential susceptibility observed in strong fields. It is necessary to discuss two possible causes of such a discrepancy.

One of them may be due to the fact that the liquid-nitrogen temperature at which we performed the measurements is close to the temperature  $T_S$  of the magnetic transformation due to the transition of the canted structure (CS) into a ferromagnetic structure, and we were therefore unable to observe the phase interchange CS—F—P (paramagnetism), which occurs in this case, since our measurements pertained entirely to the ferromagnetic region. These assumptions, however, are not justified, since the neutron-diffraction investigations<sup>[2]</sup> and the theory<sup>[1]</sup> indicate that in the indicated system there is only one alloy,  $x = 0.5$ , in which a direct CS—P (canted structure—paramagnetism) transition occurs at  $T = 234^\circ\text{K}$ ; at liquid-nitrogen temperature we should therefore have observed a large differential susceptibility in this alloy in strong fields. Nonetheless, our measurements did not confirm this.

As to the second cause, it may be more trivial and connected with the fact that our samples were not identical with those investigated in<sup>[2]</sup>, in spite of the fact that the alloys were prepared in accordance with the same procedure, developed by Lotgering and Gorter<sup>[5]</sup>. The dependence of the magnetic properties of chromium telluride and of its derivatives on the heat treatment, which causes different degrees of vacancy ordering, has been pointed out in the literature many times<sup>[4,12,13]</sup>. To verify the influence of this factor, we have performed additional measurements of the alloys ( $x = 0, 0.1, 0.2, 0.5$ ), which were heat-treated as follows: annealing for 53 hours at 400°C in evacuated quartz ampoules, followed by slow cooling in the oven. It turned out that heat treatment leads to a decrease in magnetization and to a lowering of the Curie temperature, but the form of the functional dependence of  $\sigma$  on  $H$  in strong fields remains unchanged. By way of an example, Fig. 7 shows the magnetization isotherms of two CrTe<sub>0.5</sub>Sb<sub>0.5</sub> samples—the initial one (curve 1) and after

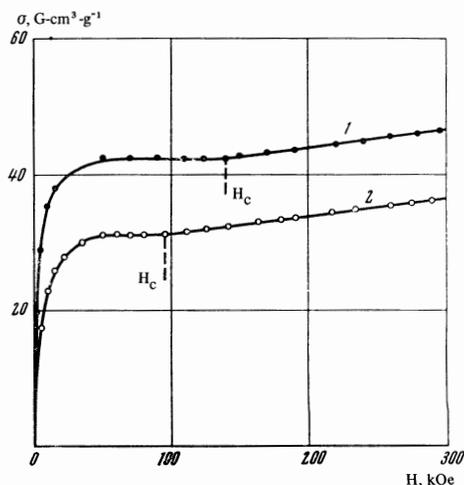


FIG. 7. Magnetization curves of CrTe<sub>0.5</sub>Sb<sub>0.5</sub> at  $T = 77^\circ\text{K}$ : 1—initial state, 2—annealing for 53 hours at 400°C.

<sup>1)</sup> An expression for the torque in the case of triangular noncollinear structures was obtained by Jacobs [1]. He has also demonstrated experimentally the absence of magnetization saturation in strong fields in a number of compounds having similar structures.

heat treatment (curve 2), measured at liquid-nitrogen temperature in pulsed fields up to 300 kOe. It is seen from the plots that the saturation magnetization decreases by approximately 25% and the critical field  $H_C$ , starting with which the magnetization begins to grow in strong fields, changes, but the general character of the change of  $\sigma$  with changing field remains the same.

On the basis of all the foregoing we arrive at the conclusion that a general idea of the magnetic phase diagram of  $\text{CrTe}_{1-x}\text{Sb}_x$  solid solutions can be obtained by starting from measurements of the magnetization at 77°K in strong pulsed magnetic fields and by determining the character of the magnetic transformations. These data make it possible to subdivide the entire system of solid solutions into four concentration regions, corresponding to different magnetic phases: 1—ferromagnetic ( $0 \leq x \leq 0.2$ ), 2—ferrimagnetic ( $0.2 < x \leq 0.5$ ), 3—noncollinear structure ( $0.6 < x < 0.9$ ), 4—antiferromagnetic structure ( $0.9 \leq x \leq 1$ ). In the composition interval  $0.5 < x \leq 0.6$  there are two magnetic phases with different magnetic structures—ferrimagnetic and noncollinear. The magnetic phase diagram constructed in accordance with the indicated subdivision is shown in Fig. 8, where the line of the Curie points is determined from the results of our measurements, the Neel points are taken from<sup>[2,5,14]</sup>, and the low-temperature transformation point  $T'_S$  of the  $\text{CrTe}_{0.3}\text{Sb}_{0.7}$  alloy was determined approximately from the temperature dependence of the magnetization, measured in a field of 8 kOe.

Let us examine the distinguishing features of each of the separated regions of the magnetic phase diagram.

The results enable us to assume that CrTe and solid solutions close to it in composition are ferromagnetic. This is confirmed by the following: a) the magnetization saturation, b) the large value of  $\xi$  corresponding to the magnetic transformation of ferromagnets at the Curie point, and c) the much better agreement, compared with ferrites, between the values of the magnetic moment determined from paramagnetic measurements and from the saturation magnetization<sup>2)</sup>.

Alloys with concentration  $x$  in the approximate interval from 0.25 to 0.5 (inclusive) can be classified as ferrimagnetic. They satisfy the thermodynamic theory of magnetic transformations, and the character of the variation of their magnetization near the Curie point is exactly the same as that of ferrites. The considerable decrease of the magnetic moment can be attributed to the presence of several magnetic sublattices with different magnetizations. In this case, the model of a ferrimagnetic crystal with three magnetic sublattices<sup>[15]</sup> makes it possible to explain the change of the magnetization in strong pulsed magnetic fields. If it is assumed that the exchange interaction of one of the sublattices,  $M_1$ , with the remaining two,  $M_2$  and  $M_3$ , which are coupled in antiparallel fashion, is much weaker than the interaction between the sublattices  $M_2$  and  $M_3$ , then sufficiently strong external magnetic fields  $H_C$  can dis-

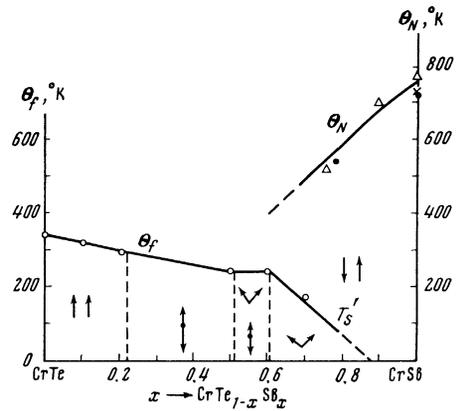


FIG. 8. Magnetic phase diagram of the  $\text{CrTe}_{1-x}\text{Sb}_x$  alloys. O—our data, ●—[<sup>2</sup>], X—[<sup>14</sup>], Δ—[<sup>5</sup>].

rupt the relatively weak exchange-coupling forces between the sublattice  $M_1$  and the antiparallel-coupled sublattices  $M_2$  and  $M_3$ . In this case an increase of the magnetization with increasing field will be observed on the  $\sigma(H)$  curve at  $H > H_C$ , after saturation had been reached. The calculations of Gusev and Pakhomov<sup>[15]</sup> have shown that in this region of fields,  $\sigma$  increases linearly with increasing magnetic field intensity, and  $d\sigma/dH$  does not depend on the temperature.

As seen from curves 1 and 2 of Fig. 2, the change of the magnetization of  $\text{CrTe}_{0.5}\text{Sb}_{0.5}$  agrees qualitatively with the scheme described above. It can be assumed that the change of  $H_C$  following heat treatment is due both to the changes in the sublattice magnetizations due to the vacancy redistribution, and to the change of the exchange parameters. We note that the Curie temperature of this alloy decreases by 42° as a result of heat treatment.

Alloys with larger Sb concentration ( $x = 0.7$  and  $0.8$ ) are characterized by the absence of saturation of the magnetization at 77°K in strong pulsed magnetic fields, thus indicating the presence of a noncollinear magnetic structure, which becomes antiferromagnetic with increasing temperature. It is quite important that the temperature of this transition ( $T'_S$ ) cannot be determined accurately on the basis of magnetic measurements. Experiment shows<sup>[3]</sup> that its determination by means of electric measurements is likewise quite difficult.

In the composition interval  $0.5 < x \leq 0.6$  there is a heterogeneous region characterized by the coexistence of two magnetic phases with different magnetic structures. One has a noncollinear structure and is responsible for the differential susceptibility in strong magnetic fields at low temperatures. The second is ferromagnetic and determines the temperature of the magnetic transformation (the Curie point) of the entire alloy as a whole. The formation of a magnetically homogeneous state in this region of compositions is also indicated by investigations of the electric properties of these alloys. It was shown in<sup>[3]</sup> that approximately in the middle of the system ( $x \approx 0.6$ ) the resistivity  $\rho$  increases strongly, differing by approximately one order of magnitude from the values of  $\rho$  of the neighboring compositions. These changes of  $\rho$ , and also the magnetic-measurement results described above, cannot be readily reconciled with the results of the neutron-diffraction investigation<sup>[2]</sup>,

<sup>2)</sup> Whether chromium telluride is ferro- or ferrimagnetic has been long under discussion in the literature [16,17,4]. The conclusion that CrTe has a ferromagnetic structure was advanced earlier [17] by one of the present authors and was later confirmed by neutron diffraction [4].

which indicate a continuous transition from ferromagnetic to antiferromagnetic ordering with increasing  $x$  in  $\text{CrTe}_{1-x}\text{Sb}_x$ . Our data indicate more readily that there exists a boundary ( $x \sim 0.6$ ) between these two magnetically-ordered states, i.e., ferro- and ferrimagnetism on one side of the phase diagram, and antiferromagnetism and noncollinear magnetic structure produced on its basis on the other.

In order to verify whether the occurrence of the noncollinear spin ordering in this system is due to the mechanism of the double exchange, let us analyze the data obtained by us for the alloys  $0.6 < x < 0.9$  on the basis of the de Gennes theory<sup>[1]</sup>. The main premises of this theory reduce to the following. The noncollinear, canted magnetic structure, consisting of basal planes that are ferromagnetically ordered and canted relative to one another, is the result of competition between double exchange and the antiferromagnetic superexchange interaction. The energy of such a system is determined by the sum of the energies  $E_{\text{ex}}$  of the antiferromagnetic superexchange and  $E_{\text{d}}$  of the double exchange:

$$E_{\text{ex}} = NzJS^2(2\cos^2(\theta/2) - 1), \quad E_{\text{d}} = -Nxzb\cos(\theta/2). \quad (3)$$

Here  $xN$  is the concentration of the current carriers participating in the double exchange,  $N$  the number of magnetically-active ions per unit volume,  $z$  the number of nearest neighbors of the magnetically active ions in adjacent layers,  $b$  the transport integral,  $J$  the parameter of super-exchange antiferromagnetic interaction, and  $S$  the spin quantum number. The angle  $\theta$  between the directions of the magnetic moments in adjacent basal planes increases with increasing  $x$ , and its magnitude is determined by the expression

$$\cos\left(\frac{\theta}{2}\right) = \frac{ax}{4}, \quad a = \frac{b}{|J|S^2}. \quad (4)$$

At low temperatures there are a small spontaneous moment ( $M_{\text{S}}$ ) and a nonzero differential susceptibility in strong fields; they are respectively equal to

$$M_{\text{S}} = \frac{Ibx}{4|J|S^2}, \quad (5)$$

$$\kappa = \frac{I^2}{4|J|zS^2N}, \quad (6)$$

where  $I/2$  is the magnetization of one sublattice and  $\kappa$  is the differential susceptibility per unit volume. Relations (3)–(6) were obtained by de Gennes for the case  $b \gg k\theta N$ , i.e., it was assumed that the width of the conduction band is relatively large and the carriers fill only the bottom of the band. According to estimates by Anderson<sup>[18]</sup>, the transport integral in the double-exchange mechanism should be of the order of 0.1–0.3 eV for oxides of transition metals (the Anderson-Hasegawa double exchange<sup>[19]</sup>), and  $b \sim 1$  eV for systems similar to  $\text{CrTe}_{1-x}\text{Sb}_x$ . It should be noted that an experimental determination of  $b$  is of interest in itself, for the purpose of verifying the theory of double exchange.

As already mentioned, neutron-diffraction investigations of  $\text{CrTe}_{1-x}\text{Sb}_x$  solid solutions made it possible to determine the change of the angle  $\theta$  between the magnetic sublattices with increasing concentration  $x$ . This has made it possible to calculate the ratio  $\alpha \approx 5$ . Measurements

**Table II.** Specific ( $\sigma_{\text{S}}$ ) and volume ( $M_{\text{S}}$ ) spontaneous magnetizations and differential susceptibilities of the  $\text{CrTe}_{1-x}\text{Sb}_x$  alloys in strong magnetic fields (150–300 kOe)

Alloy	Magnetization at $H = 0$		Differential susceptibility		$4M_{\text{S}}/I$
	$\sigma_{\text{S}}$ , G-cm <sup>3</sup> /g	$M_{\text{S}}$ , g	$\chi \cdot 10^5$	$\kappa \cdot 10^4$	
$\text{CrTe}_{0.4}\text{Sb}_{0.6}$	28.0	165.20	4.53	2.67	1.16
$\text{CrTe}_{0.3}\text{Sb}_{0.7}$	17.4	102.66	4.81	2.84	0.72
$\text{CrTe}_{0.2}\text{Sb}_{0.8}$	3.3	19.47	3.58	2.11	0.136

in strong magnetic fields make it possible to determine this ratio by an independent method. We have attempted to estimate qualitatively the value of  $\alpha$  for alloys with large Sb content ( $0.6 \leq x < 0.9$ ), since the existence of a canted magnetic structure could be assumed for them. To this end we use the spontaneous-magnetization values determined by linear extrapolation of the  $\sigma(H)$  curves at  $T = 77^\circ\text{K}$  to a zero field, and the values of the differential susceptibility in strong fields. These data are given in Table II in addition to the values of  $4M_{\text{S}}/I = \alpha\kappa$ , calculated under the condition that the magnetization of one sublattice ( $I/2 = 285$  G) is determined by the effective magnetic moment of the chromium ion  $M_{\text{eff}} = gS$ , assuming complete “quenching” of the orbital angular momentum at  $S = 3/2$  and  $g = 2$ .

The relation  $4M_{\text{S}}/I = f(x)$  could be approximated by a straight line, from which  $\alpha$  could be found to equal  $\sim 5.1$ . This result agrees with<sup>[2]</sup>. It was assumed in the calculations that the magnetization of one sublattice and the integral of the antiferromagnetic superexchange interaction do not change with  $x$ , in agreement with expression (6) and the obtained experimental data, which indicate that the low-temperature differential susceptibility changes little with  $x$ . The values of the parameter  $|J|$ , calculated for the  $\text{CrTe}_{1-x}\text{Sb}_x$  alloys from formula (6), lie in the range  $(27-35) \times 10^{-16}$  erg, which agrees with  $|J| = 33 \times 10^{-16}$  erg of the antiferromagnetic alloy CrSb, as determined from the Neel temperature<sup>3)</sup>. On this basis we could estimate the transport integral  $b \sim 0.02$  eV, which turned out to be much smaller than the theoretical estimates<sup>[1,18]</sup>.

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<sup>3)</sup>In the calculation by means of formula (6) we used the following data:  $S = 3/2$ ,  $N = 2.46 \times 10^{22}$  cm<sup>-3</sup>, and  $z = 2$ . The specific gravity of the alloy was assumed to be 5.9 g/cm<sup>3</sup> for all the compositions.

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214