

absorption coefficients, etc.

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Effect of high pressure and doping on the Curie temperature in chromium telluride

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Results of an experimental investigation of the effect of hydrostatic pressure (up to 12 kbar) on the Curie temperature T_c of solid $\text{CrTe}_{1-x}\text{X}_x$ solutions ($\text{X} = \text{Se}, \text{Sb}$) are presented for concentration values x up to 0.5. The baric coefficients $\gamma = T_c^{-1}(dT_c/dP)$ of all alloys investigated are negative. However the form of $\gamma(x)$ is determined by the doping element X and accordingly by the nature of the compression, which can be either isotropic ($\text{X} = \text{Se}$) or anisotropic ($\text{X} = \text{Sb}$). Possible mechanisms of exchange spin coupling in the investigated alloys are discussed on the basis of the obtained data. It is concluded that two types of exchange interactions coexist in chromium telluride: 1) indirect exchange of localized electrons via the anion and 2) interaction between collectivized electrons in the narrow 3d band, and the energy of this interaction determines the Curie temperature.

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INTRODUCTION

The Curie temperature of chromium telluride depends very strongly on the variation of the interatomic distances, a fact resulting apparently from singularities in the exchange spin coupling in chalcogenides of 3d transition metals, the nature of which is not clear to this day. These questions have been intensively investigated in recent years both experimentally (see, e. g., ^[1,2]) and theoretically. ^[3,4]

In earlier studies^[5,6] of chromium telluride at high pressures, attempts were made to distinguish between the influences exerted on the Curie temperature by two factors—volume changes and substitution of one anion for another by formation of solid solutions $\text{CrTe}_{1-x}\text{X}_x$, where $\text{X} = \text{S}, \text{Se}, \text{or Sb}$. It was established there that the Curie point T_c decreases with decreasing interatomic distance and the value of the derivative $dT_c/dV = 3 \cdot 10^{25} \text{ deg/cm}^3$ does not depend on the manner in which the lattice has been compressed, by hydrostatic pressure or by introducing the impurity S, Se, or Sb. The present paper is a continuation of the aforementioned research.

To answer the question why the Curie temperature of chromium telluride changes under hydrostatic compression and what is the mechanism that produces the exchange interactions in this alloy, we investigated two systems of solid solutions, $\text{CrTe}_{1-x}\text{Se}_x$ and $\text{CrTe}_{1-x}\text{Sb}_x$, which differ from each other in that when the tellurium atoms are replaced by selenium the hexagonal lattice becomes almost isotropically compressed (the alloys $\text{CrTe}_{1-x}\text{Se}_x$), and when the solid solutions $\text{CrTe}_{1-x}\text{Sb}_x$ are produced, an anisotropic change in volume takes place as a result of the strong decrease of the hexagonal axis.

We present in this paper the results of an experimental investigation of the magnetization, the crystal-lattice parameters, and the baric characteristics of the Curie temperature (dT_c/dP and $\gamma = T_c^{-1}(dT_c/dP)$) of the alloys $\text{CrTe}_{1-x}\text{Se}_x$ and $\text{CrTe}_{1-x}\text{Sb}_x$ in the concentration interval $0 \leq x \leq 0.5$. On the basis of the obtained data we discuss the previously proposed mechanisms of the exchange interactions in chromium tellurides^[1,4] and conclude that the observed regularities in the variation of the Curie temperature with pressure can be explained on the basis of the ferromagnetism of the collectivized electrons.

MEASUREMENT PROCEDURE AND RESULTS

The solid solutions $\text{CrTe}_{1-x}\text{Se}_x$ and $\text{CrTe}_{1-x}\text{Sb}_x$ were synthesized by heating thoroughly mixed powders of the initial chemically-pure elements in evacuated quartz ampoules. The thermal synthesis regime was analogous to that described by Lotgering and Gorter.^[1] Special attention was paid to the phase composition of the alloys, for which purpose an x-ray-diffraction analysis and a microstructural phase analysis were performed, the results of which demonstrated the investigated alloys are single-phase, with the exception of the stoichiometric compound CrTe . In the latter case, besides the fundamental phase CrTe , there were also small amounts of Cr , in which case the x-ray-patterns revealed a slight broadening of the large-angle lines: (203), (121), (114), and (122). The x-ray patterns were obtained from powder samples in an RKD-57 camera by an asymmetric method in chromium and copper radiation. All the interference lines on the x-ray patterns obtained at room temperatures were indexed on the basis of a hexagonal lattice of the type NiAs (space group $P6^3/\text{mmc}$). The interference line (001) of the superstructure of the ordered vacancies was not observed by us. The parameters of the unit cell a and c were determined by graphically extrapolating the plots of

$$a, c = f(\varphi), \quad \varphi = \left(\frac{\cos^2 \theta}{\sin \theta} + \frac{\cos^2 \theta}{\theta} \right)$$

to zero. The errors were $\pm 0.002 \text{ kX}$ for the parameter a and $\pm 0.005 \text{ kX}$ for the parameter c .

The magnetic measurements were performed with the aid of a pendulum magnetometer^[7] in fields up to 16 kOe. The Curie temperature was determined by the method of thermodynamic coefficients, developed by Belov.^[8] The change ΔT_c of the Curie temperature as a result of hydrostatic compression was determined from the shift of the plots of the temperature dependence of the magnetization $\sigma(T)$, measured at different pressures in a field $H = 8 \text{ kOe}$. The method used by us to produce pressure by "locking and piston"^[9] has caused the pressure in the chamber to decrease monotonically with decreasing temperature. This circumstance decreased the accuracy of ΔT_c , since the $\sigma(T)$ curves were plotted not at constant P , but in a certain interval ΔP (the pressure drop was $\sim 3 \text{ kbar}$ on going from room temperature to 77°K). As a result, ΔT_c was determined in the fol-

TABLE I. Curie temperature T_c , lattice parameters, and unit-cell volume of the alloys $\text{CrTe}_{1-x}\text{X}_x$ (where $\text{X} = \text{Se}$ or Sb).

x	T_c, K	a, kX	c, kX	c/a	$V, (\text{kX})^3$
X=Se					
0.07	308	3.975	6.229	1.567	85.23
0.10	293	3.961	6.220	1.570	84.51
0.25	220	3.913	6.197	1.584	82.16
0.30	201	3.897	6.189	1.588	81.40
0.40	137	3.868	6.158	1.592	79.79
0.50	100	3.850	6.110	1.587	78.42
X=Sb					
0	342	3.997	6.230	1.559	86.20
0.10	319	3.993	6.171	1.542	85.07
0.20	299	3.998	6.113	1.528	84.58
0.35	280	4.005	6.040	1.508	83.90
0.50	249	4.015	5.931	1.471	82.78

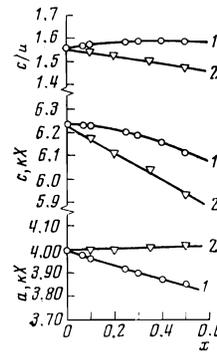


FIG. 1. Dependence of the crystal-lattice parameters of the alloys $\text{CrTe}_{1-x}\text{X}_x$ on x : curves 1—for $\text{X} = \text{Se}$, curves 2—for $\text{X} = \text{Sb}$.

lowing manner. On the $\sigma(T)$ curves measured in a field 8 kOe at atmospheric pressure, we marked the magnetization value corresponding to the true Curie temperature determined by the method of thermodynamic coefficients. At other pressures, we picked out on the $\sigma(T)$ curves the same values of σ , and the temperatures corresponding to them were taken to be T_c . In some cases the shift of the Curie point with changing pressure was determined from the plots of the temperature dependences of the resistance $R(T)$ and of the galvanomagnetic effect $\Delta R(T)/R$; the obtained values of ΔT_c determined by different methods were in good agreement with one another.

We used in the measurement a high-pressure chamber made of the titanium alloy VTZ-1 and described by us previously.^[9] The pressure-transmitting medium was a mixture of transformer oil and isopentane. The temperature in the chamber was measured with a copper-constantan thermocouple and was maintained constant within $\pm 0.5^\circ$, while the pressure was determined from the electric resistance of a wire-wound manganin manometer accurate to 50 bar.

MEASUREMENT RESULTS

Table I and Figs. 1 and 2 show the results of the measurements of the Curie temperature, the lattice parameters, and the unit-cell volume (V) of the solid solutions $\text{CrTe}_{1-x}\text{Se}_x$ and $\text{CrTe}_{1-x}\text{Sb}_x$ as functions of the selenium or antimony contents. It is seen from these data that the alloys $\text{CrTe}_{1-x}\text{Se}_x$ and $\text{CrTe}_{1-x}\text{Sb}_x$ behave differently, namely, both parameters a and c decrease simultaneously with increasing selenium concentration, as a result of which the lattice becomes almost isotropically compressed with a very small change in the ratio c/a (curve

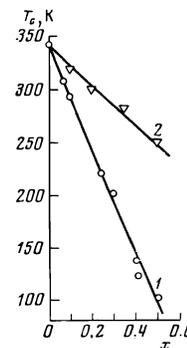


FIG. 2. Change of Curie temperature of the $\text{CrTe}_{1-x}\text{X}_x$ alloys as a function of x : curve 1—for $\text{X} = \text{Se}$; 2—for $\text{X} = \text{Sb}$.

1 of Fig. 2). To the contrary, as seen from curve 2 of Fig. 2, replacement of the tellurium atoms with antimony is accompanied by an anisotropic compression of the volume as a result of the strong decrease of the hexagonal axis. The parameter a remains practically unchanged, but the ratio c/a decreases with increasing Sb concentration.

From Fig. 3, which shows the plots of $T_c(x)$, it is seen that the rate of change of the Curie point of chromium telluride doped with antimony is somewhat lower than in the case of selenium doping. In the present study we have investigated solid solutions in the concentration interval $0 \leq x \leq 0.5$, corresponding to the region where these alloys are magnetically single-phase. It was reported on the basis of neutron-diffraction data^[10] that a continuous transition from the ferromagnetic CrTe to the antiferromagnetic CrSb takes place in these systems in an ordered fashion, with formation of intermediate noncollinear magnetic structures. Later, however, in an investigation of the resistivity ρ as a function of x in a series of $\text{CrTe}_{1-x}\text{Sb}_x$ solid solutions it was noted^[11] that approximately at the midpoint of the system $x \approx 0.5$, a magnetically two-phase state is produced, connected with the existence of ferro- and antiferromagnetically ordered regions. The possibility of formation of magnetically heterogeneous states in analogous systems was indicated also by Takei *et al.*^[12] and it was noted that the treatment of the neutron-diffraction data, on the basis of which the magnetic state in the region $x \approx 0.5$ can be interpreted as having a two phase structure, or else as having the noncollinear so called canted antiferromagnetic structure, is ambiguous.

In Fig. 3, using the data of Table I, we plotted T_c as a function of the unit-cell volume for all the investigated alloys, and marked on the same plots the values of T_c obtained for CrTe at hydrostatic pressures up to 12 kbar^[6] and quasi-hydrostatic pressures up to 30 kbar.^[13] It is seen from Fig. 4 that all the points fit quite well a single straight line $T_c(V)$, calculated by using the experimental data: $dT_c/dP = -6.2$ deg/kbar, and using the compressibility^[1] $\chi = 24.5 \times 10^{-7}$ cm²/kg. Attention is called to the fact that when the Te atoms in the chromium tellurides are replaced by antimony, only small changes of T_c and V are possible within the limits of the homogeneous solid solution. Thus, for example, at the boundary of the single-phase region the volume of the $\text{CrTe}_{0.5}\text{Sb}_{0.5}$ unit cell is equal to $V = 82.78$ (kX)³ and $T_c = 249$ °K, thus imitating the state of the CrTe alloy at

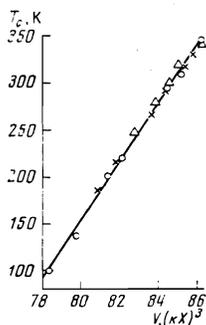


FIG. 3. Dependence of the Curie temperature on the unit-cell volume of the alloys $\text{CrTe}_{1-x}\text{X}_x$: \circ — $\text{X}=\text{Se}$, Δ — $\text{X}=\text{Sb}$, \times —change of V under the influence of hydrostatic pressure.

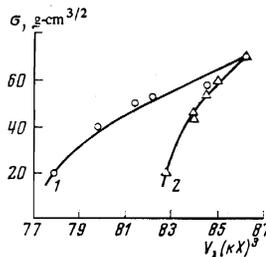


FIG. 4. Change of specific magnetization ($H = 8$ kOe) as a function of the unit-cell volume for the alloys $\text{CrTe}_{1-x}\text{Se}_x$ (curve 1) and $\text{CrTe}_{1-x}\text{Sb}_x$ (curve 2).

pressures ~ 16 kbar. When the chromium telluride is doped with selenium ($\text{CrTe}_{0.5}\text{Se}_{0.5}$), a larger decrease in the volume of the unit cell is possible, and consequently also in the Curie temperature: V decreases to 78.42 (kX)³ and T_c decreases to 100 °K, corresponding to the action of a pressure 30–35 kbar. It is most important that investigations of chromium telluride carried out with the same samples at pressures up to 50 kbar by the electron magnetic resonance method^[13] have shown that in the pressure region 25–30 kbar, at 100 °K, the resonance line characteristic of the ferromagnetic state vanishes, possibly as a result of the onset of a new magnetic phase transition at high pressures.

Figure 4 shows the variation of the specific magnetization of the solid solutions $\text{CrTe}_{1-x}\text{Se}_x$ (curve 1) and $\text{CrTe}_{1-x}\text{Sb}_x$ (curve 2) as a function of the unit-cell volume V of the crystal. The measurements of σ were performed at atmospheric pressure and $T = 77$ °K in an external field 8 kOe. It is seen from the plots that the magnetization, in contrast to T_c , is determined principally by the concentration of the doping elements, while the influence of the volume factor plays here a secondary role. Namely, in the case $\text{X}=\text{Sb}$ (curve 2, Fig. 4) in the interval $0 \leq x \leq 0.35$ the volume decreases insignificantly, from 86.2 to 83.9 (kX)³, but the magnetization decreases strongly with increasing x , the change of σ amounting to 25 G-cm³/g. With further increase of x , to 0.5 , a sharp drop of σ takes place, corresponding to the boundary of the magnetic single-phase region of these alloys. If the doping is with selenium in the interval $0 \leq x \leq 0.4$ (curve 1, Fig. 4), approximately the same decrease takes place, $\Delta\sigma = 30$ G-cm³/g, but V changes in this case much more (from 86.2 to 79.8 (kX)³). It is seen from the diagram that the alloys $\text{CrTe}_{1-x}\text{X}_x$ ($\text{X}=\text{Se}$ or Sb) with concentration $x = 0.5$ have the same values of the magnetization ($\sigma = 20$ G-cm³/g) at different V , i.e., the change of σ is determined principally by the number of tellurium atoms replaced by the doping element.

Table II and Figs. 5, 6, and 7 show the result of measurements of the Curie temperature, the derivatives dT_c/dP , and the baric coefficients $\gamma = T_c^{-1}(dT_c/dP)$ of all the measured alloys. The investigated pressure interval is indicated for each case.

Figure 5 shows the results of the measurement of T_c as a function of the pressure for the alloys $\text{CrTe}_{1-x}\text{Se}_x$. Analogous data were obtained also for the system $\text{CrTe}_{1-x}\text{Sb}_x$. In all cases, the Curie point decreases linearly with increasing pressure, but the values of dT_c/dP and the baric coefficients of the Curie point γ change

TABLE II. Baric characteristic of the Curie temperature of the alloys $\text{CrTe}_{1-x}\text{X}_x$ (where X=Se or Sb).

x	T_c, K	$-\frac{dT_c}{dP}, \frac{\text{deg}}{\text{kbar}}$	$\frac{1}{T_c} \frac{dT_c}{dP} \cdot 10^{-3}, \text{bar}^{-1}$	Pressure interval, kbar
X=Se				
0.10	293	-6.4 ± 0.4	-2.18	0.001-7.7
0.25	220	-7.1 ± 0.8	-3.25	0.001-12.5
0.30	201	-8.1 ± 0.6	-4.03	0.001-8.0
0.40	137	-8.8 ± 0.8	-6.35	0.001-7.0
0.50	100	-8.7 ± 1	-8.67	0.001-4.0
X=Sb				
0	342	-6.2 ± 0.3	-1.81	0.001-12.0
0.10	319	-6.0 ± 0.3	-1.88	0.001-10.0
0.20	299	-5.4 ± 0.4	-1.86	0.001-11.4
0.35	280	-5.0 ± 0.5	-1.79	0.001-10.5
0.50	249	-4.2 ± 0.3	-1.69	0.001-12.1

differently, depending on which of the elements, Se or Sb, enters in the alloy, and consequently an isotropic (X=Se) or anisotropic (X=Sb) change of volume is produced by the doping.

From the plots of Fig. 6 it is seen that the quantities $-dT_c/dP$ increase with increasing x for the alloys $\text{CrTe}_{1-x}\text{Se}_x$ and decrease for the alloys $\text{CrTe}_{1-x}\text{Sb}_x$. The baric coefficients γ do not change with changing antimony concentration, and in the alloys $\text{CrTe}_{1-x}\text{Se}_x$ they increase with increasing x (Fig. 7). These data point to the presence of a correlation between the change of the derivatives dT_c/dP and the character of the compression of the crystal upon doping. Taking into consideration the results of the described x-ray diffraction investigations above, it can be stated that the relation $-\gamma(x) \sim x$ takes place in the case of an isotropic decrease of the volume, while $-\gamma(x) = \text{const}$ in the case of anisotropic compression of the lattice.

DISCUSSION OF RESULTS

Analyzing the data obtained in the present paper, we must consider two questions: first, why is the Curie temperature of the solid solutions $\text{CrTe}_{1-x}\text{X}_x$ determined only by the volume and does not depend on which of the anions (Te, Se, or Sb) enters in the alloy, while the magnetization, to the contrary, is determined principally by the number of Te atoms replaced by the doping element; second, why does the character of the compression (isotropic or anisotropic change of volume) not influence the $T_c(V)$ dependence, but this factor manifests itself clearly in the volume dependence of the baric derivatives of the Curie temperature, $dT_c(V)/dP$ and $\gamma(V)$.

The observed irregularities can be explained by as-

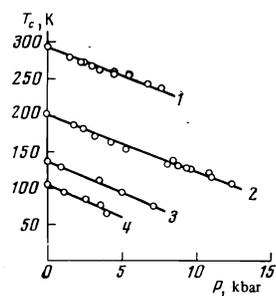


FIG. 5. Dependence of the Curie temperature on the pressure for the alloys $\text{CrTe}_{1-x}\text{Se}_x$: curve 1— for $x=0.1$, 2— for $x=0.3$, 3— for $x=0.4$, and 4— for $x=0.5$.

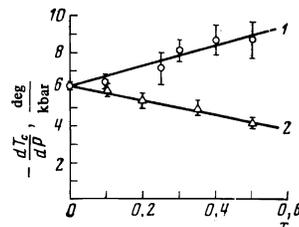


FIG. 6. Dependence of the derivative dT_c/dP of the $\text{CrTe}_{1-x}\text{X}_x$ alloys on the concentration x : curve 1— for X=Se; curve 2— for X=Sb.

suming that in the investigated alloys there coexist localized and collectivized $3d$ electrons and accordingly there exist at least two different types of exchange interactions: a) indirect superexchange of localized electrons via the anion, as is evidenced by the dependence of the magnetization on the anion that determines the character of the exchange interaction of the nearest environment, and b) interaction in the collectivized-electron system, with an energy that exceeds energies of the existing interactions and determines the Curie temperature.

It appears that it is precisely the last fact which is the reason why the onset of the antiferromagnetically interacting pairs Cr-Se-Cr or Cr-Sb-Cr does not lead, with increasing concentration x of the ternary alloys $\text{CrTe}_{1-x}\text{X}_x$ to a stronger decrease of the Curie temperature in comparison with the shift of T_c with changing pressure in chromium telluride. In addition, there is likewise no appearance of the most characteristic feature of the superexchange, connected with the spatial directivity of the pattern, as determined by the mutual arrangement of the cations and anions.

It should be noted that the idea of the coexistence of localized or collectivized $3d$ electrons in chalcogenides of transition metals was first advanced by Goodenough.^[4] From the experimental data obtained by us, however, it is seen that some of Goodenough's qualitative considerations, connected with the postulate that direct antiferromagnetic exchange takes place along the hexagonal axis, do not hold true for chromium tellurides. Namely, the largest value of dT_c/V should be observed in the alloys $\text{CrTe}_{1-x}\text{Sb}_x$, where the entire change of the volume is due to the decrease of the c axis, but our experimental data refute these assumptions. Nonetheless, the main idea of dividing the $3d$ electrons into two groups with different types of exchange interaction seems to be correct.

Starting from the foregoing, the change of the Curie temperature of chromium telluride under the influence of pressure and doping can be qualitatively understood on the basis of the general premises of the model of collectivized electrons. According to this model, T_c is

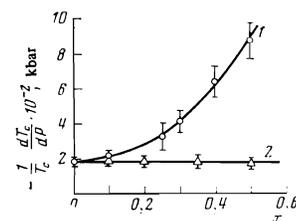


FIG. 7. Change of baric coefficients of the Curie temperature of the alloys $\text{CrTe}_{1-x}\text{X}_x$ as a function of x : curve 1— for X=Se, 2— for X=Sb.

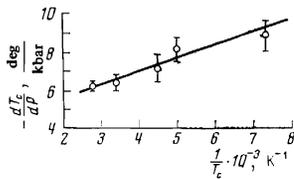


FIG. 8. Dependence of the derivative dT_c/dP on $1/T_c$ of the alloys $\text{CrTe}_{1-x}\text{Se}_x$.

determined in first-order approximation by the product of two quantities $N(E_F)$ and U_{eff} , i. e., by the density of states on the Fermi surface and the effective energy of the interelectron interaction. Consequently, the sign of dT_c/dP will be determined by the manner in which each of the indicated parameters changes separately under the influence of the pressure. This question was investigated theoretically for Ni by Lang and Ehrenreich^[14] and most fully for weak band ferromagnets of the ZrZn_2 type by Wohlfarth.^[15] Later, on the basis of the theory of band ferromagnetism, alloys of 3d-transition metals with a distinguished narrow 3d-energy band was considered, the number of electrons in which remains unchanged under hydrostatic compression and the character of the $N(E)$ curve remains the same. The following relations were obtained^[16,17]:

$$\frac{\partial T_c}{\partial P} = \frac{5}{6} \kappa \left[2T_c - \frac{B}{T_c} \right], \quad (1)$$

$$B = U_{\text{eff}} T_F^2 / U, \quad (2)$$

$$U_{\text{eff}} = U / (1 + \alpha U W^{-1}), \quad (3)$$

$$\bar{U} = U_{\text{eff}} N(E_F),$$

$$\alpha = \frac{1}{2} W \int_{E_F}^w N(E) \frac{dE}{E}, \quad (4)$$

$$T_F^2 = \frac{6}{k_B \pi^2} \left[\left(\frac{N'(E_F)}{N(E_F)} \right)^2 - \frac{N''(E_F)}{N(E_F)} \right]. \quad (5)$$

In these expressions, κ is the compressibility, U is the interelectron interaction without allowance for correlations, and T_F is the effective degeneracy temperature, which is connected with the density of the electronic states $N(E_F)$ and its derivatives $N'(E_F)$ and $N''(E_F)$ on the Fermi surface. The interelectron interaction is determined by Eq. (3) in accordance with Kanamori^[18] while α does not depend on the width W of the band.

It follows from (1) that in alloys with high Curie temperatures the principal role is played by the term proportional to T_c , and the sign of the derivative dT_c/dP should be positive, whereas in alloys with low Curie temperatures the term $-B/T_c$ predominates and leads to negative values of dT_c/dP . In this case one should expect a parabolic variation of T_c with increasing P and the presence of a critical pressure $P_{\text{cr}} = T_c(0)^2/2B$ at which the ferromagnetism should vanish.

The measurement results shown in Table II and in Fig. 8, which shows also the dependence of $-dT_c/dP$ on $1/T_c$ for $\text{CrTe}_{1-x}\text{Se}_x$ alloys, offer evidence that the relation (1) describes correctly the experimental data obtained with isotropic compression of the crystal. It is obvious that the linear dependence $T_c(P)$ obtained in our experiment is due to the small interval of the investigated pressure region. An estimate of the critical pressure connected with the vanishing of the band ferro-

magnetism in chromium telluride yields a value $P_{\text{cr}} \sim 27.6$ kbar, which is close to a magnetic phase transition observed in experiments on magnetic resonance.^[13]

By considering the experimental data in light of all the foregoing, we can conclude that the cause of the strong lowering of T_c with pressure in chromium telluride is the decrease of the density of states $N(E_F)$ as a result of the broadening of the 3d band when the crystal is compressed and the Fermi level shifts in the energy band.

Another result connected with the experimentally established correlation between the change of dT_c/dP and the character of the compression seems to indicate that the baric derivatives of the Curie temperature are determined not so much by the quantity $N(E_F)$ as by the shape of the state-density curve. Namely, the discrepancy between the theory (relation (1)) and experiment are observed in the case of anisotropic compression of the doped crystal ($-dT_c/dP \sim T_c$), when the probability of distorting the band structure and the form of the $N(E)$ curve is large. It is possible that in this case an important role will be assumed by the change of U_{eff} , since we do not know how the interelectron interaction varies as a function of the structure of the d band and of the pressure.

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