

# Quasi-Two-Dimensional Ferromagnetism in $\text{Cr}_2\text{Te}_3$ and $\text{Cr}_5\text{Te}_8$ Crystals

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The crystal structure of  $\text{CrTe}$  suggests two-dimensional magnetism in the plane perpendicular to the  $c$  axis. It is consistent with the temperature dependence of the  $g$  factor and EPR lines width observed. In the present paper we investigate the  $\text{CrTe}$  alloy which has been prepared by melting of the powdered  $\text{Cr}_2\text{Te}_3$  in evacuated quartz ampoule at the temperature 1600 K. ESR spectra were recorded using X-band (9.4 GHz) spectrometer provided with gas nitrogen cryostat. The shape of the EPR line depends strongly on the temperature. In the vicinity of the room temperature the lines become very wide and weak or disappear completely. At lower temperatures the shape of the lines approaches the Dyson function. The asymmetry of lines is attributed to the strong exchange interactions as well as to the semimetal electrical conductivity. Above the room temperature the shape of spectra is characteristic for the paramagnetic phase. Results were addressed based on the model of critical spin fluctuations in two-dimensional Heisenberg magnet proposed by Eremin et al.

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## 1. Introduction

Two-dimensional (2D) ferromagnets and antiferromagnets are extensively studied theoretically and experimentally due to their future applications in spintronic devices. 2D magnetic behaviour has been observed and studied in such systems as: manganites [1],  $\text{Cu(en)(H}_2\text{O)}_2\text{SO}_4$  single crystals [2], self-organized assemblies of nanoclusters [3], ultrathin films of diselenides (see [4] and refs. therein), films of  ${}^3\text{He}$  [5] or nanolayers of  $\text{ZnO}$  [6].

Chromium chalcogenides are alternative — with respect to diluted magnetic semiconductors — materials for spintronic devices production. Cr-chalcogenides reveal hexagonal NiAs-type crystal structure. Crystals of this type have attracted considerable attention due to their layered structure which determines peculiar electrical and magnetic properties of such materials (e.g. new FeAs type of high temperature superconductivity [7]). Moreover, in Cr-chalcogenides, alterations of the number or the arrangement of Cr-vacancies, as well as the substitution of metal ions, lead to different classes of crystals of the  $\text{Cr}_{1-x}(\text{Te, S, Se})$  type, with a wide range of many fascinating electrical and magnetic properties. Crystals of these compounds, such as  $\text{Cr}_{1-x}\text{Te}$ , exhibit antiferromagnetism, non-collinear spin structures, or itinerant electron magnetism [8]. The substitution of Cr atoms with Ti or V creates new properties such as spin-glass [9, 10].

Some chalcogenides (e.g.  $\text{Cr}_{1-x}\text{Te}$  or  $\text{Cr}_{1-x}\text{Se}$ ) exhibit metastable zinc-blende structure [11], which allows for the growth of zinc-blende solid solution semiconductor compounds, with their properties similar to those of diluted magnetic semiconductors, e.g.  $\text{CrMnTe}$  [12],  $\text{ZnCrTe}$  [13],  $\text{ZnCrSe}$  [14].

In this paper, we investigate, by ESR, the  $\text{Cr}_{1-x}\text{Te}$  alloy with a hexagonal structure of NiAs.

The crystal structure of  $\text{Cr}_{1-x}\text{Te}$  suggests two-dimensional magnetism in the plane perpendicular to the  $c$  axis. It is consistent with the temperature dependence of the  $g$  factor and the EPR lines width observed.

## 2. Experimental

Samples have been prepared by melting of the powdered  $\text{Cr}_2\text{Te}_3$  (Alfa Aesar, 99.5%) in evacuated ( $\approx 10^{-5}$  mbar) quartz ampoule at temperature 1600 K. The ampoule was gradually heated from the room temperature up to 1600 K for 1 h. Then it was kept at 1600 K for half an hour and then it was cooled step by step to the room temperature for 2 h. X-ray powder diffraction patterns were obtained at the room temperature on Bruker XRD D8 Advance with DAVINCI diffractometer using  $\text{Cu K}_\alpha$  radiation  $\lambda = 1.54060$  Å. Samples were placed on phistage with 20 rpm rotation. Diffraction reflections were collected in the range  $2\theta = 20\text{--}120^\circ$ . Phase analysis was studied using Bruker Era software with PDF-2 2012 database as well as the Fityk package equipped with standard ASTM diffraction data cards. Two predominant phases  $\text{Cr}_5\text{Te}_8$  and hexagonal  $\text{Cr}_2\text{Te}_3$  were determined from the diffraction pattern. The intensive line at  $2\theta = 25.67^\circ$  was attributed to cubic (zinc blende) phase of  $\text{Cr}_2\text{Te}_3$  with lattice constant 6.464 Å. Both phases,  $\text{Cr}_5\text{Te}_8$  and hexagonal  $\text{Cr}_2\text{Te}_3$ , are of NiAs type. Very small discrepancy between values of the parameter  $x$  ( $x = 0.33$  and  $x = 0.37$  for  $\text{Cr}_2\text{Te}_3$  and  $\text{Cr}_5\text{Te}_8$ , respectively) should be noticed. The crystal similarity of both phases determine the same magnetic properties studied in the present paper.

The ESR spectra were recorded using the X-band (9.4 GHz) spectrometer provided with gas nitrogen cryostat (Oxford Instruments). The spectra were measured in the range temperature of 150–350 K.

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### 3. Results

The shape of the ESR line depends strongly on the temperature. There are two regions where the spectra become very wide and weak or disappear completely. These temperatures are 325–315 K and below 200 K.

In Fig. 1a the fitting of the experimental lines at  $T = 240$  K and 350 K to the Dysonian and the Lorentz function is presented. The dependence of the  $g$ -factor on temperature is shown in Fig. 1b.

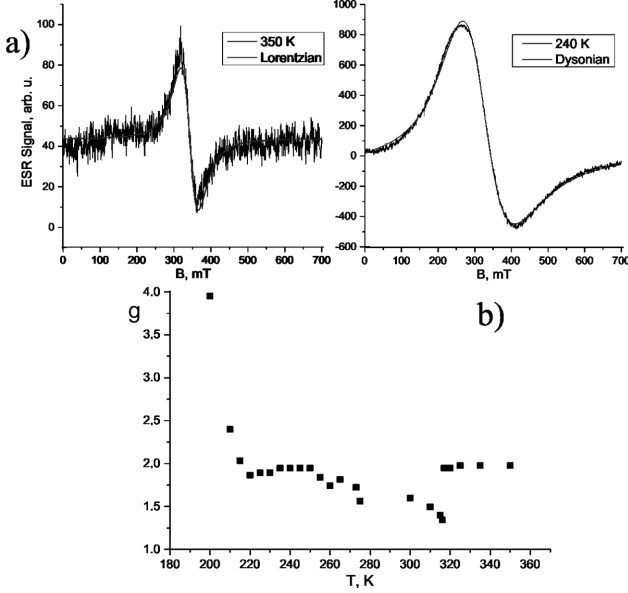


Fig. 1. The fitting of the experimental lines at  $T = 240$  K and  $T = 320$  K to Dysonian and Lorentzian respectively (a), dependence of the  $g$ -factor on temperature (b).

At lower temperatures, the shape of the lines approaches the Lorentz function. The asymmetry of the lines is also visible, which is attributed to the strong exchange interaction, as well as to the semimetal electrical conductivity. Above the room temperature, the shape of the spectra is characteristic for the paramagnetic phase. At this range of temperature, the ESR lines are stable with respect to their position, as well as to the width. Moreover, they are relatively narrow.

### 4. Discussion and conclusions

Crystals of  $\text{Cr}_2\text{Te}_3$  have a layered structure of the NiAs type [15]. It consists of perfect hexagonal metal layers (Cr) separated with a Te layer and a metal layer with a high number of vacancies (e.g. the vacancy Cr layer consists of 25% empty Cr sites). In the model of the structure presented in Fig. 2, the metal layer (the black balls) is fully occupied by Cr. In the hexagonal layer, each Cr atom is surrounded by six nearest Cr neighbors (Fig. 2). This layer is surrounded by two other layers on the left and on the right side: these are the nonmagnetic Te layers. The elementary cell of  $\text{Cr}_2\text{Te}_3$  is magnified in

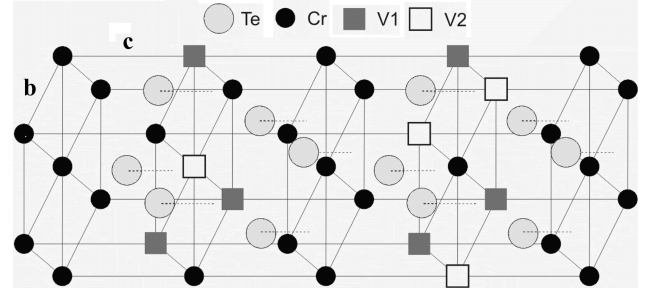


Fig. 2. Model of  $\text{Cr}_2\text{Te}_3$  and  $\text{Cr}_5\text{Te}_8$  crystal structure (v1 and v2 denotes two types of Cr vacancies).

the  $z$  direction due to different positions of vacancies in the neighbouring vacancy layers [15] (Fig. 2).

The crystal structure of  $\text{Cr}_2\text{Te}_3$  prefers two-dimensional magnetic properties. Due to the low dimensionality, specific features are also observed in the ESR measurements. It has been commonly known that the pure two-dimensional Heisenberg model with nearest-neighbour ferromagnetic interactions does not undergo a phase transition. This result was supported by Takahashi [16, 17], who — within the framework of the modified spin-wave theory — shows that in two-dimensional case, the susceptibility diverges according to

$$\chi = CT^l \exp\left(\frac{\alpha}{T}\right), \quad (1)$$

where  $C$ ,  $l$ ,  $\alpha$  are constants depending on approximation.

However, Stanley and Kaplan [18] addressed the problem and suggested the possibility of a phase transition for the quasi two-dimensional Heisenberg model. In the case of a weak magnetic interaction  $J_\perp$  of the two-dimensional layer with parallel neighbor layer, the magnetic susceptibility at the temperature  $T > T_C$  was derived within the random-phase-approximation [18]:

$$\chi(\mathbf{q}, \omega) = \frac{\chi_{2d}(\mathbf{q})}{1 - J_\perp(\mathbf{q}) \chi_{2d}(\mathbf{q})} \frac{i\gamma(\mathbf{q}, \omega)}{\omega + i\gamma(\mathbf{q}, \omega)}, \quad (2)$$

where  $J_\perp$  is the ferromagnetic exchange between spins from different layers,  $\gamma(\mathbf{q}, \omega)$  is the damping function of the spin fluctuations,  $\chi_{2d}(\mathbf{q})$  is the static spin susceptibility in the layer.

For the Heisenberg ferromagnet on a triangular lattice, Eremin et al. [19] obtained the following formulae for the two-dimensional susceptibility and for the Curie temperature of the system, respectively:

$$\zeta_{2d}(T) = \frac{1}{\sqrt{6}} \left[ \frac{J_\parallel}{T} \right]^{-\frac{1}{2}} \exp\left(\frac{8\pi\sqrt{3}J_\parallel S}{T}\right), \quad (3)$$

$$k_B T_C \approx \frac{16\sqrt{3}J_\parallel S^2}{\ln \frac{6\sqrt{3}J_\parallel S}{J_\perp}}, \quad (4)$$

where  $S$  is the spin of ions forming lattice.

The magnetic susceptibility, which is proportional to the ESR intensity  $I_{\text{ESR}}$ , was determined from the formula [20]:

$$I_{\text{ESR}} = I\Delta B^2 (1 + \alpha^2)^{0.5}, \quad (5)$$

where  $I = A + B$  is the amplitude of  $dP/dB$  and  $\alpha$  is the ratio of dispersion to absorption. The coefficient  $\alpha$  has been determined from the asymmetry coefficient  $\alpha' = B/A$ ,  $\alpha = 1 - \alpha'$ , where  $A$  and  $B$  are the amplitudes of the low field and the high field halves of the signal, respectively. Dependence of  $I_{\text{ESR}}$  on temperature is presented in Fig. 3.

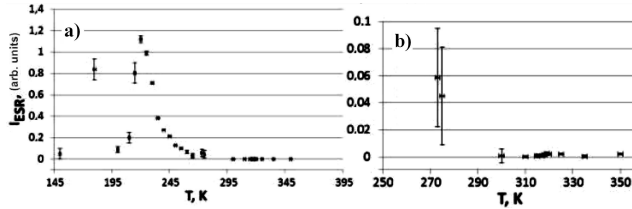


Fig. 3. Temperature dependence of ESR susceptibility.

The bulk ferromagnetism transition at 210 K is visible in Fig. 3a very well. The second phase transition is observed at 295 K (Fig. 3b). ESR susceptibility is not sensitive considerably to this transition, while various Cr layer may have opposite ferromagnetic magnetization in the  $z$  direction. However, this transition is better visible in temperature dependence of linewidth (Fig. 4).

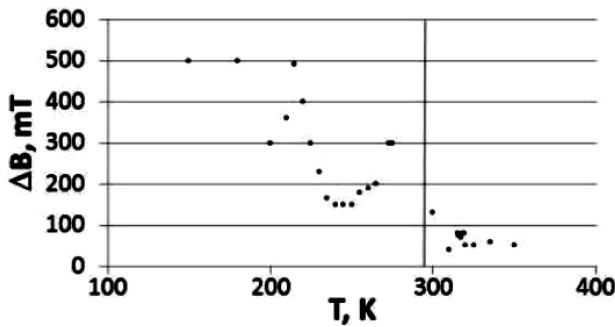


Fig. 4. Temperature dependence of linewidth (two-dimensional transition is visible at 295 K).

## 5. Conclusions

ESR measurements in  $\text{Cr}_2\text{Te}_3$  crystal reveal two types of magnetic transitions. The first (at 295 K) relates to quasi-isolated two-dimensional Cr planes in crystal structure of the NiAs type. The second (at 210 K) follows from bulk ferromagnetism. Two-dimensional behavior of Cr spins is better visible in the temperature dependence of width of ESR lines comparing to such dependence of ESR susceptibility.

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