EXCHANGE INTEGRALS FOR THE FIRST AND SECOND COORDINATION SPHERES IN THE NEW SPINEL SERIES Cu$_{1-x}$ZnxCr$_2$Te$_4$ (where $x = 0.00, 0.01, 0.02$)

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EXCHANGE INTEGRALS FOR THE FIRST AND SECOND COORDINATION SPHERES IN THE NEW SPINEL SERIES Cu$_{1-x}$Zn$_x$Cr$_2$Te$_4$ (where $x = 0.00, 0.01, 0.02$)

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Abstract. - The exchange integrals for the first and second coordination spheres of the spinels under study were determined. The former turned out to be positive and the latter-negative. Thus, the ferromagnetic structure of these spinels is a result of the competition of the ferro- and antiferro-interactions.

Introduction

The compounds CuCr$_2$X$_4$ (where $X = S, Se, Te$) are ferromagnets with the relatively high Curie temperature (equal to 420, 460, 360 K for $X = S, Se, Te$, respectively) and with the type p metallic conductivity [1]. These properties differ clearly from the properties of the chalcogenide spinels (CdCr$_2$Se$_4$, HgCr$_2$Se$_4$ etc.) [2], which are semiconductive ferromagnets with the Curie temperature considerably lower than room temperature. From the investigations of several authors as well as from our investigations [3] it follows, that there are Cu$^+$ ions which are responsible for these interesting physical properties of the chromium spinels. For instance, insertion of a Cu$^+$ ion in place of a Zn$^{2+}$ ion in the compounds Cu$_x$Zn$_{1-x}$Cr$_2$Se$_4$ causes a change of valence of a chromium ion from 3+ to 4+ resulting in a strong ferromagnetic coupling between the magnetic moments localized on these ions (3 $\mu_B$ for $\text{Cr}^{3+}$ and 2 $\mu_B$ for $\text{Cr}^{4+}$). The aim of this paper is the determination of the exchange integrals for the first and the second coordination spheres in the isostructural compounds Cu$_x$Zn$_{1-x}$Cr$_2$Te$_4$.

Experimental

Polycrystalline samples of the compounds Cu$_x$Zn$_{1-x}$Cr$_2$Te$_4$ [4] were obtained for $x = 0.00, 0.01, 0.02$ by heating substrates in the quartz ampules evacuated down to the pressure of $10^{-5}$ Pa at 923 K. These compounds crystallize with the space group ofFd3m. The magnetic investigations were carried out in the fields with the induction $B$ up to 14 T and in the temperature range of 4.2-650 K with the aid of both the induction and the Faraday method. It turned out, that the magnetic saturation moment at 4.2 K decreases with the increase of Zn concentration in the sample; the ferromagnetic Curie temperature $T_C$ decreases as well, whereas the paramagnetic Curie-Weiss temperature $\theta_{\text{C-W}}$ increases. The representative $\mu$ vs. $B$ curves at 4.2 K and 295 K are presented in figure 1 while both the $\sigma$ vs. $T$ and $\chi_M$ vs. $T$ curves are shown in figure 2. As it is concluded in [4] all the samples under study reveal the metallic character of the conductivity. The most important experimental results are listed in table I.

![Fig. 1. The dependence of the magnetic moment $\mu$ on the induction of the external magnetic field for Cu$_{0.99}$Zn$_{0.01}$Cr$_2$Te$_4$.](image1)

![Fig. 2. The dependence of the specific magnetization $\sigma(T)$ and the magnetic susceptibility $\chi_M(T)$ for Cu$_{0.99}$Zn$_{0.01}$Cr$_2$Te$_4$.](image2)
Table I. — The most important experimental data and the calculated exchange integrals for Cu$_{1-z}$Zn$_z$Cr$_2$Te$_4$.

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The determination of the exchange integrals

In order to determine the exchange integrals for the first and the second coordination spheres, i.e. $J/k$ and $K/k$ the theory presented in [5] was utilized. It turns out, that in the MFA for the spinels containing the magnetic moments only in the octahedral sublattice it is possible to write down the equations linking the magnetic phase transition temperatures with the exchange integrals. The exchange integrals $J$ and $K$ were calculated from the equations:

$$\theta_{c-w} = \frac{5}{2k} (6J + 36K)$$  \hspace{1cm} (1)

$$T_c = \frac{5}{2k} (2J - 4K)$$  \hspace{1cm} (2)

which were derived from the equations taken from [5]. The obtained results are presented in table I.

Discussion

The exchange integrals determined for the first coordination sphere are positive and big (compare e.g. spinels CdCr$_2$S$_4$, CdCr$_2$Se$_4$ and HgCr$_2$Se$_4$ with the values of the exchange integrals equal to 11.8 K, 14 K and 15.8 K, respectively [2]). For the second coordination sphere the exchange integrals are negative and smaller. As a result of the competition of these two interactions a ferromagnetic order is established. It is well known [2], that in the chalcogenide spinels a superexchange interaction is responsible for the ordering of the magnetic moments localized on the chromium ions. A question then arises, what is the additional exchange interaction mechanism (apart from the superexchange interactions), that is responsible for the values of the exchange integrals in the compounds Cu$_{1-z}$Zn$_z$Cr$_2$Te$_4$ which are several times bigger comparing to the chalcogenide spinels. This additional exchange interaction mechanisms is most probably the double exchange interaction just as in the case of the spinels Cu$_{1-z}$Zn$_z$Cr$_2$Se$_4$ [3]. The investigations to solve this problem are in progress.

Acknowledgment

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