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The Features of Thermomagnetic Properties of Solid Solutions $Cu_{1-x}In_xCr_2S_4$

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Abstract

The field and concentration dependences of the thermomagnetic parameters of spinel solid solutions $Cu_{1-x}In_xCr_2S_4$ have been researched. Anomalous magnetoresistance and magnetothermal-e.m.f. have been observed depending on the composition and strength of magnetic field related to the presence of weak hopping transitions in solid solutions $Cu_{1-x}In_xCr_2S_4$ ($0 \le x \le 0.3$). Negative magnetoresistance has been observed in weak magnetic field intensities which indicates the presence of inelastic scattering while indirect exchange interaction occurring with low hopping transition. A quadratic dependence of magnetothermal-e.m.f. on the saturation magnetization has been revealed as well that qualitatively conforms to the theoretical calculations carried out for the ferromagnetic semiconductors.

Keywords: *Solid solutions, Ferromagnetic semiconductors, Thermomagnetic parameters, Magnetoresistance, Magnetothermal-e.m.f.*

1. Introduction

The simultaneous presence of electron, phonon and localized magnetically ordered centres gives a special interest to the study of spinel semiconductors [1]. Solid solutions $Cu_{1-x}In_xCr_2S_4$ also belong to the class of similar materials making the study of their thermomagnetic properties current one.

2. Experimental part

The synthesis of the alloys was carried out from the elements: Cu – electrolytic, Cr of Erkh mark, sulfur «OSCh» and indium In-000. Technological mode of obtaining alloys of the studied system was similar to the methods cited in [2], with a slight difference of synthesis temperature (±303÷323 K) and homogenization annealing. At interaction of copper, indium, chrome and sulfur some sharply expressed heat effects were observed on the thermograms what testifies to intensive proceeding of reaction between these elements with isolation of a great volume of heat. Homogenizing annealing was carried out at ~1073 K within 300 h. Afterwards such thermal treatment the samples were brought to a powdery state and compressed, then endured within 10 days at temperature of 873 K. Consequently, the process of thermal treatment of samples had two stage character. The thermomagnetic properties were studied on the compressed samples of the parallelepiped form in the 80-703 K range.

3. Results and discussion

The results of our investigations have shown that abnormal behaviours are available in the changes of the values of magnetoresistance $\left(\Delta\rho/\rho\right)$ and magnetothermal-e.m.f. $\left(\Delta\alpha/\alpha\right)$ of solid solutions $Cu_{1-x}In_xCr_2S_4$, where $0 \le x \le 0.35$, associated with the presence of weak hopping transitions in these crystals. With the increase of indium amount in the solid solution composition $Cu_{1-x}In_xCr_2S_4$ the disappearance is observed depending $\Delta\rho/\rho$ on the composition of such anomalies. Fig. 1 shows the concentration dependences of the specific (ρ_0) and magnetoresistance $\left(\Delta\rho/\rho\right)$ of studied solid solutions at 300K and magnetic field strength 11 kOe.

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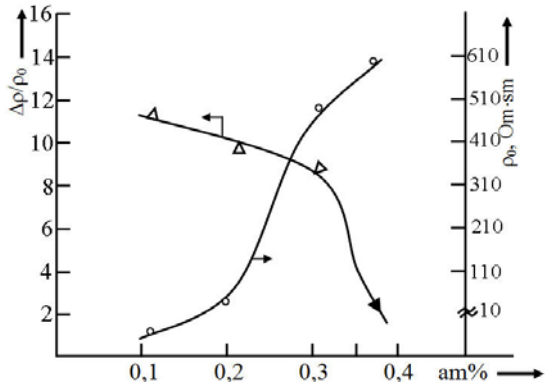

Fig. 1. Concentration dependence of specific (ρ_0) and magnetoresistance ($\Delta\rho/\rho$) of $Cu_{1-x}In_xCr_2S_4$ solid solutions at 300K and magnetic field H=11 kOe

As it is evident, with the increasing of the amount of In^{3+} ions in the composition the magnetic resistance decreases, and when x=0.35 it reaches a minimum value and the specific resistance is maximum in this composition. On the other hand, the weakening of the magnetic ordering is carried out simultaneously with the increase in specific resistance. Fig. 2 shows the dependence of the magnetoresistance on magnetic field strength of a solid solution with $Cu_{0,90}In_{0,10}Cr_2S_4$ composition at two temperatures. Negative magnetoresistance is observed at 317 K and low values of the magnetic field strength. This anomalous behaviour $\Delta\rho/\rho_0$ along with weak hopping transitions in this composition, availability of inelastic scattering of charge carriers while direct exchange interaction [3].

A similar anomaly is not observed at relatively high (~365K) temperature. The measurements showed that magnetoresistance in $Cu_{0,80}In_{0,20}Cr_2S_4$ composition with the magnetic field strength of 9 kOe has a value of ~0.0314, and for $Cu_{0,70}In_{0,30}Cr_2S_4$ composition $\Delta\rho/\rho_0$ value is ~0.0029.

The research of magnetothermal-e.m.f. of solid solutions $Cu_{1-x}In_xCr_2S_4$ showed the quadratic dependence of this parameter on low magnetic field strength. However, saturation is observed at stronger (higher) magnetic field strengths when there is a disappearance (cancellation) of the domain structure in $\Delta\alpha/\alpha \sim f(H)$ dependences (Fig. 3).

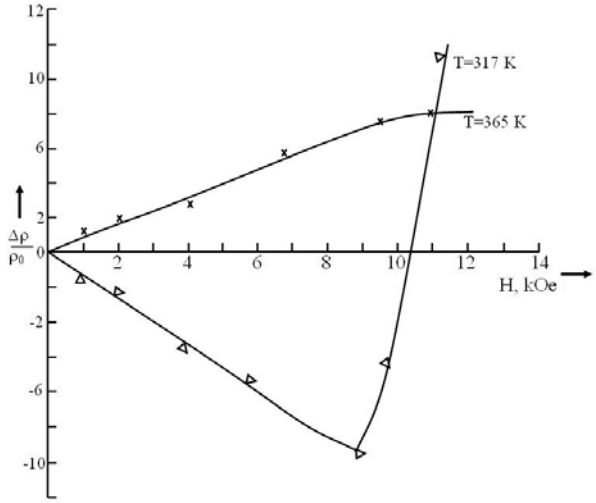

Fig. 2. Dependence of the magnetoresistance of solid solution with $Cu_{0,90}In_{0,10}Cr_2S_4$ composition on the magnetic field strength at different temperatures

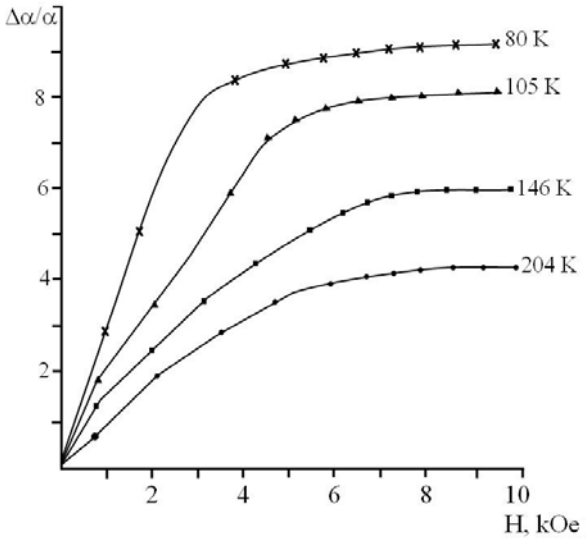

Fig. 3. Dependence of magnetothermal-e.m.f. on the magnetic field strength of solid solution $Cu_{0,80}In_{0,20}Cr_2S_4$ at different temperatures

The spontaneity of Nernst-Ettingshausen effect and saturation magnetizations (M_S) according to found values are defined after extrapolation of the curves of $\Delta\alpha/\alpha \sim f(H)$ dependence to the zero point (N=0). The quadratic dependence $\Delta\alpha/\alpha$ on this parameter is defined. Constructed dependence

$\Delta\alpha/\alpha \sim f\left(M_S/M_{S_0}\right)$ is shown in Fig. 4 which is characterized by a quadratic nature of change. We note that the quadratic nature of the dependencies $\Delta\rho/\rho_0 \sim f(M_S)$ and $\Delta\alpha/\alpha \sim f(M_S)$ qualitatively conforms to the theoretical charting of the same parameters [4]. The absolute values of thermo-magnetic parameters do not exceed 18 and 7 per cent which indicates the presence of a weak exchange interaction and localized magnetic moment in the researched solid solutions.

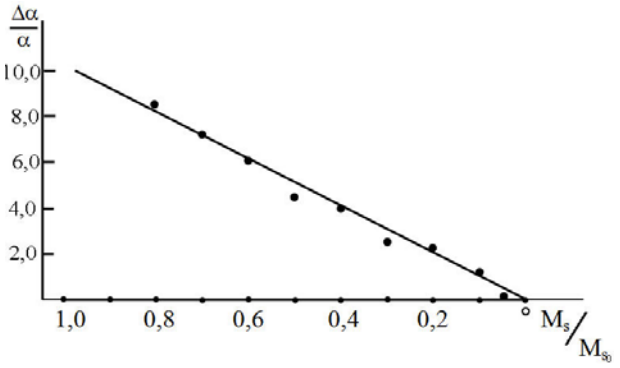

Fig. 4. The dependence of spontaneous magnetothermal-e.m.f. on relative magnetization for the composition of the solid solution $Cu_{0,80}In_{0,20}Cr_2S_4$

However, with an increase of the amount of indium in the composition, the indirect exchange interaction process somewhat enhances. For a more clear understanding of the mechanism of this process, let us turn to the temperature dependences of magnetothermal-e.m.f. and carrier mobility coefficient [5] where it is determined that the scattering of charge carriers in solid solutions $Cu_{1-x}In_xCr_2S_4$ at low temperatures derive mainly from the impurity ions of indium. According to the neutrality principle and magnification of magnetothermal-e.m.f. coefficient under the influence of magnetic field on $\Delta\alpha$ value we can assume that the charge carriers can scatter at the same time due to charging impurity atoms. Taking this into account, according to the value of the scattering coefficient by the following well-known formula [6] for certain formulations of the researched solid solutions the value of Fermi energy:

$$\alpha = \frac{1}{3e}\pi^2 k^2\left(r+\frac{3}{2}\right)\frac{T}{E_F}.$$

It was found that the value of Fermi energy increases too with the increase of the amount of indium in composition. The values of the effective mass of the charge carriers have been calculated subject to the parabolic band structure according to the formula $\frac{m^*}{m_0} = h^2(3\pi p)^{\frac{2}{3}}$ are given in [7]. It has been found that $m^*=0.7m_0$ subject to total polarization of charge carriers. For weak polarization of the carriers between the split small subbands of the valence area $m^*=0.5m_0$.

Fig. 5 shows the concentration dependence of magnetothermal-e.m.f. $Cu_{1-x}In_xCr_2S_4$ solid solutions. As we can see, this option changes its sign with a change in the composition. Such a change may be an indicator of changes in the scattering mechanism of charge carriers. These factors and the nature of interaction of charge carriers with magnetic structural half-systems lead to anomalous behaviours of kinetic coefficients of solid solutions $Cu_{1-x}In_xCr_2S_4$.

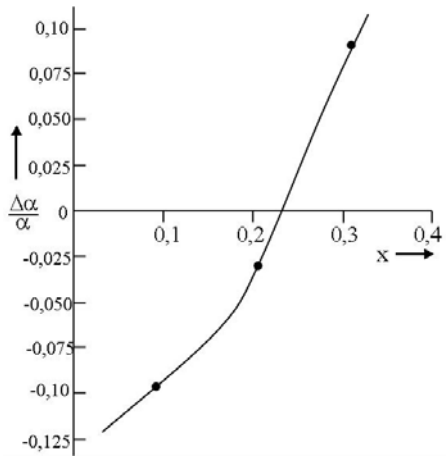

Fig. 5. Concentration dependence of magnetothermal-e.m.f. of solid solution alloys $Cu_{1-x}In_xCr_2S_4$

4. Conclusions

Thus, from the study of thermomagnetic properties it was established that with increasing of the amount of In^{3+} ions in the $Cu_{1-x}In_xCr_2S_4$ solid solutions the magnetic ordering weakens and due to

increase the indirect exchange interaction hopping transitions are activated.

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