HgCrCdSe as an element of new heterostructure HgCrCdSe/HgMnTe

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By using the EPR method we show that the introduction of the fourth component as the cadmium (Cd) into the solid solution $Cr_xHg_{1-x}Se$ allows for increasing the transition temperature of magnetic ordering in this compound. The resistance, magnetoresistance, and current-voltage characteristics of new heterojunctions HgCdCrSe/HgMnTe are measured as a function of temperature and external magnetic field. The appearance of nonlinear plot on the reverse branch of experimental current-voltage characteristic at a temperature below the temperature of magnetic ordering can be explained by the appearance of spin-polarized current.

С привлечением метода ЭПР показано, что введение в твердый раствор $Cr_xHg_{1-x}Se$ четвертого компонента в виде кадмия (Cd) позволяет повысить температуру перехода данного соединения в фазу магнитного упорядочения. Электросопротивление, магнитосопротивление и вольт-амперные характеристики новых гетеропереходов HgCdCrSe/HgMnTe измерены в зависимости от температуры и внешнего магнитного поля. Наличие нелинейного участка на обратной ветви экспериментальных вольт-амперных характеристик при температурах ниже температуры магнитного упорядочения можно объяснить появлением спин-поляризованного тока.

1. Introduction

The existing theoretical models of spin electronic devices point out the great promise perspectives of the given scientific research [1, 2]. In this connection there is a need to create heterojunctions which allow for implementation of spin electronic devices as well as to develop the experimental methods of spin-polarized transport investigations. To implement spin devices with high performance it is necessary to solve several problems, among which are the following: ensuring a high degree of spin polarization of charge carriers and high operating temperatures, as well as an efficient transfer of polarized current carriers and preservation of this polarization in the nonmagnetic components of suitable heterojunctions [3, 4]. By using diluted magnetic semiconductors (DMS) as magnetic components of such heterojunctions one can meet most of aforementioned requirements since by varying the composition of each component one can independently change their electric and magnetic characteristics. Special attention is paid to increase the Curie temperature of DMS up to room temperature [5, 6]. In addition, to minimize the scattering related to possible impurities and dislocations, it is necessary to find a heteropair with the good agreement of their lattice constants. It is these conditions will help to preserve the coherence of the polarized electrons flow without affecting the length of their free path [7].

The purpose of this paper is to apply DMS CdHgCrSe as spin injector and to

carry out the experimental investigations of new heterostructure HgCdCrSe/HgMnTe in terms of getting a spin-polarized transport at temperatures higher than nitrogen ones.

All the samples under test were grown at Chernivetsky National University by Prof.S. Yu.Paranchich.

2. Experimental

In our previous studies we have shown that ferromagnetic ordering is observed in the solid solutions $Cr_xHg_{1-x}Se$ for the content of chromium $0.03 < x < 0.1$ at the temperature $T = 106$ K due to presence of spinel clusters $HgCr_2Se_4$ [8]. We hypothesized that introduction into the solution of the fourth component of cadmium (Cd) will increase the transition temperature of ferromagnetic ordering due to the formation of two clusters as $CdCr_2Se_4$ and $HgCr_2Se_4$ having the structure of normal spinel with the Curie temperatures T_C equal to 130 K and 106 K, respectively. Based on the foregoing, the crystal of solid-solution $Cd_xHg_{1-x-y}Cr_ySe$ was grown with the initial content of Cd ($x = 0.4$) and Cr ($y = 0.1$). We investigated a series of samples cut from the central region of the ingot.

Microanalysis of CdHgCrSe samples performed on scanning electron microscope ZEISS EVO 50XVP system with a combined system of the energy-dispersive analysis of INCA ENERGY 450 and structural analysis performed on the instrument HKL Channel 5, have shown that they have a heterogeneous composition. As an example, in Fig. 1 an electronic image of the part of surface of one of the samples is presented. Detailed analysis of the sample shows that the percentage of Cd, Hg, Cr and Se in the dark areas corresponds to the solid solution CdHgCrSe (Table 1, the spectra Fig 1–4). The bright areas correspond to the solid solution CdHgSe (Table 1, the spectra Fig 5, 6). Thus, in contrast to our assumptions about
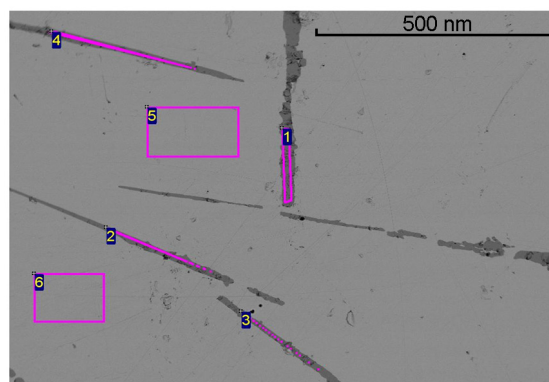

Fig. 1. Electronic image of the surface part of the sample CdHgCrSe.

the possible formation of two types of ferromagnetic inclusions in this compound there are two types of solid solutions with chromium in one of them. It should be noted that the percentage of CdHgCrSe is less than 10 %, as well as the percentage of $HgCr_2Se_4$ in the solid solutions CrHgSe [8]. In addition, for the samples under test only minor variations in the percentage of the above-mentioned solid solutions were observed from sample to sample.

3. Magnetoresonance characteristics of CdHgCrSe

Magnetoresonance properties of solid solutions CdHgCrSe were investigated by ESR method in the temperature range 77 K $< T < 300$ K.

In Fig. 2 the values of EPR line intensity of the samples cut from the central region of the ingot (samples numbering starts from the beginning of the ingot) have shown. The findings suggest that the minor changes in the concentration of chromium are observed in the transition from sample to sample.

Fig. 3 shows typical results obtained for one of the samples. Analyzing the EPR spectra the temperature dependence of pa-

Table 1. The samples composition in the areas (Note: all results are presented in weight percentages)

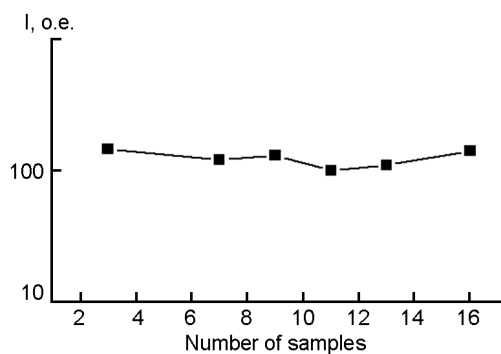| Spectrum	Cr	Se	Cd	Hg	Total
1	18.90	51.51	8.70	20.89	100.00
2	17.49	53.55	8.38	20.57	100.00
3	18.96	51.28	8.53	21.23	100.00
4	18.35	51.97	9.46	20.22	100.00
5		32.27	28.37	39.36	100.00
6		30.72	26.38	42.90	100.00
Max.	18.96	53.55	28.37	42.90	
Min.	17.49	30.72	8.38	20.22	

Fig. 2. EPR line intensity of the Cr^{3+} ions for the samples $Cd_xHg_{1-x-y}CrySe$.

rameters such as the ESR line intensity, *g*-factor and EPR line width were built. As follows from these dependences, the last two parameters of the EPR spectrum are beginning to increase sharply until nitrogen temperatures starting from the temperature $T = 157$ K. The intensity of the ESR line increases almost two orders of magnitude, *g*-factor increases from $g = 2.045$ to $g = 2.522$, and the ESR line width increases almost 8 times. The increase of the EPR line width with the temperature decrease is typical for spinel ferrites with a mixed-valence [9–11]. The temperature dependence of the EPR line intensity (Fig. 3a) obeys the Curie-Weiss law (1):

$$\chi = \frac{C}{T - T_C}, \qquad (1)$$

where C is a constant, T_C is the Curie temperature. As a result of approximation of this dependence the Curie temperature $T_C = 124$ K for four-component solution CdHgCrSe was obtained. Note that in this case an increase in the temperature of ferromagnetic ordering is observed in comparison with three-component solution HgCrSe ($T_C = 106$ K).

However, unlike the three-component solid solution $Cr_xHg_{1-x}Se$ the mechanism of ferromagnetic ordering in the samples CdHgCrSe is different. In the substitution solution like that the chromium ions usually found in the crystal lattice nodes, replacing mercury ions. Cr^{2+} ions give an electron in the conduction band and become ions Cr^{3+} [12]. The energy diagramm of semiconductors like that has an inverted structure of *s*- and *p*-bands. As the temperature decreases and approaches the Curie temperature, the bottom of the conduction band is shifted downward and the impurity
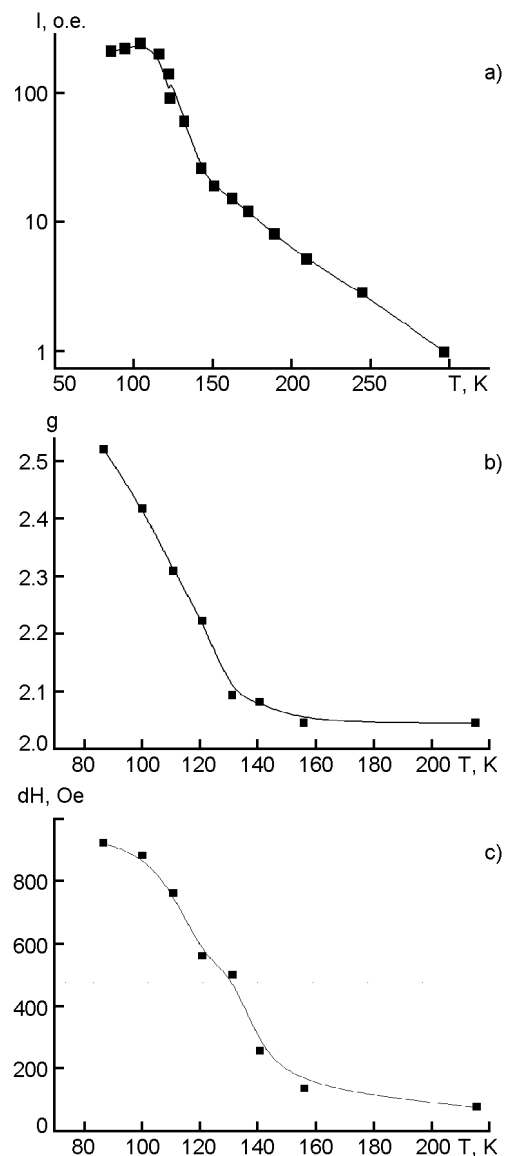

Fig. 3. The temperature dependencies of the EPR line intensity (a), *g*-factor (b), and the EPR line width (c) for the samples $Cd_xHg_{1-x-y}CrySe$.

band may overlap with the conduction band. Depending on the degree of hybridization of these bands the mobility of conduction electrons and their concentration will change.

This effect has a significant impact on the conductive and magnetic properties of semiconductors. Thus, the semiconductor CdHgCrSe under test gets a mixed valence of Cr^{2+} ($3d^4$) and Cr^{3+} ($3d^3$) ions. In this case *d*-electrons are localized in the crystal lattice nodes, and *s*-electrons are collectivized. Accordingly, if a semiconductor with Cr^{2+} ions shows a weak paramagnetism of Van Vleck type, a semiconductor with Cr^{3+} ions shows the paramagnetic properties. In

this case, with the temperature decrease the formation of ferromagnetic ordering occurs via the exchange interaction of the localized electrons with the conduction electrons, which leads to the long-range oscillating exchange coupling.

For quadratic dispersion law for conduction electrons the exchange integral has the form [13]:

$$J_{eff}(R) = -9\pi \frac{I^2}{E_F}(n_s)^2 F(2k_F R), \qquad (2)$$

where E_F — Fermi energy for the conduction electrons, n_s — conduction electron concentration, I — s–d exchange integral describing the exchange interaction between s-electron and d-electron located in the node taken as the reference point; F — Ruderman-Kittel function, k — electron wave vector.

In the framework of this model an increase of the Curie temperature up to 124 K for CdHgCrSe can be explained by an increase in the concentration of conduction electrons, since according to the results of structural analysis the concentration of chromium and, accordingly, the charge carriers concentration in CdHgCrSe higher than in HgCrSe [8].

Thus, it follows from our results, the ferromagnetic ordering takes place in the solid solution $Cd_xHg_{1-x-y}Cr_ySe$, as clearly indicate the temperature dependences of the ESR line intensity, g-factor and EPR line width.

However, in order to build the final model of ferromagnetic ordering formation in the samples $Cd_xHg_{1-x-y}Cr_ySe$ the additional research should be performed to study the mechanisms of adjustment of the band structure of such a semiconductor in the region of the phase transition as a change both in its composition and in the conduction electrons concentration.

4. Characteristics of heterojunctions HgCdCrSe/HgMnTe

The above-described semiconductor $Cd_xHg_{1-x-y}Cr_ySe$ was used as a component of new heterojunction HgCdCrSe/HgMnTe. The measurements of electrical resistivity, magnetoresistance and current-voltage characteristics of the heterojunctions were carried out at the setup for semiconductor's studying [14]. By knowing the Curie temperature for the given quaternary compound, we analyze the characteristics of the heterojunction

Table 2. Resistances of heterojunctions at $T = 300$ K

No. of the sample	Electrical resistance, Ohm	Magnetoresistance, %
1	0.6	4.9
2	0.8	8.3
3	0.4	6.8
4	5.4	9.9

HgCdCrSe/HgMnTe at the temperatures before and after it. The measurements of current-voltage characteristics were performed in the range of bias voltages $(-0.05$ V$) \le U \le (+0.05$ V$)$ at temperatures ranging from the room to the liquid nitrogen temperature with $10°$ increments using the procedure described in [15,16]. The investigated heterojunctions had different ohmic resistance at the room temperature (Table 2).

As can be seen from Table 2, the electrical resistivity of the heterojunctions No.1–No.3 at the room temperature are quite low and not significantly different among themselves (0.4 Ohm–0.8 Ohm), while the electrical resistivity of the heterojunction No.4 is significantly higher ($R = 5.4$ Ohm). Note that the temperature dependences of electrical resistivity of the heterojunctions No.1 and No.2 vary slightly with decreasing the temperature down to the liquid nitrogen one, and the electrical resistivity of the heterojunction No.3 increases noticeably (0.4 Ohm–2.1 Ohm) (Fig. 4a). In this case the temperature dependence of electrical resistivity for the heterojunction No.4 is different from previous ones (Fig. 4a), namely, there is a significant increase of the electrical resistivity starting with the temperature $T = 157$ K. It is worth noting that the abrupt changes in the EPR line characteristics of the samples HgCdCrSe start with the same temperature (Fig. 3a). By lowering the temperature down to the nitrogen one the electrical resistance of the heterojunction No.4 increases to $R = 9.6$ Ohm. This temperature dependence of the electrical resistance is typical for the DMS in their transition from the paramagnetic to the ferromagnetic phase [17].

The measured temperature dependences of magnetoresistance of the heterostructures under study has shown in Fig. 4b. The observed magnetoresistance increase with the temperature decrease is caused by an increase of the charge carriers mobility due to reducing the influence of thermal lattice
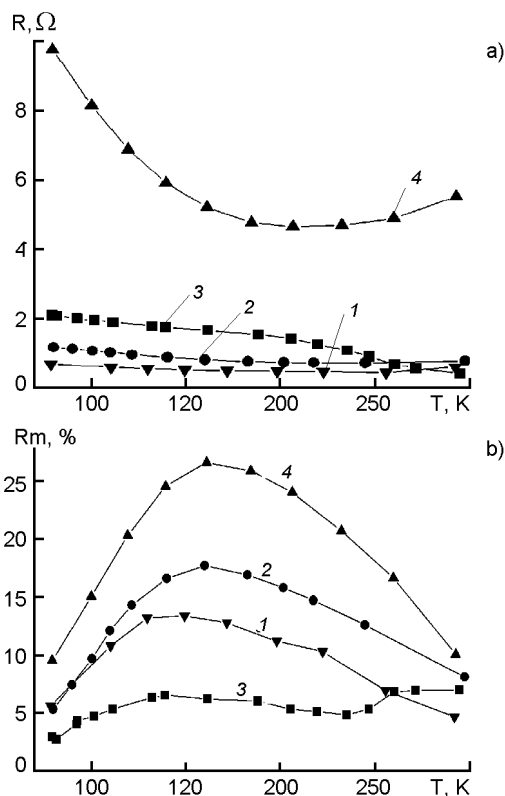
Fig. 4. Temperature dependences of the electrical resistance (a) and magnetoresistance (b) of heterojunctions CdHgCrSe/MnHgTe (H = 5000 Oe).

vibrations on the magnetic ordering. The further resistivity decrease below the temperature $T \approx 157$ K can be attributed to the weakening of charge carriers scattering by the localized magnetic ions as an establishment of the ferromagnetic order. The scattering mechanisms noted above cause the extreme character of the temperature dependence of the magnetoresistance. A similar temperature dependence of the magnetoresistance is typical to the ferromagnetic materials, in particular, for the ferromagnetic semiconductors [18]. Both components of the aforementioned heterojunctions above the Curie temperature are paramagnetic and their current-voltage characteristics are linear (Fig. 5a). At temperatures below the Curie temperature there is a non-linear part on the reverse branch of the current-voltage characteristic of the heterojunction No.4 (Fig. 5b). This kind of current-voltage characteristics have been observed previously in the structures of metal-n–HgCr$_2$Se$_4$ [19]. The explanation of our results can be carried out in the framework of the following physical model.
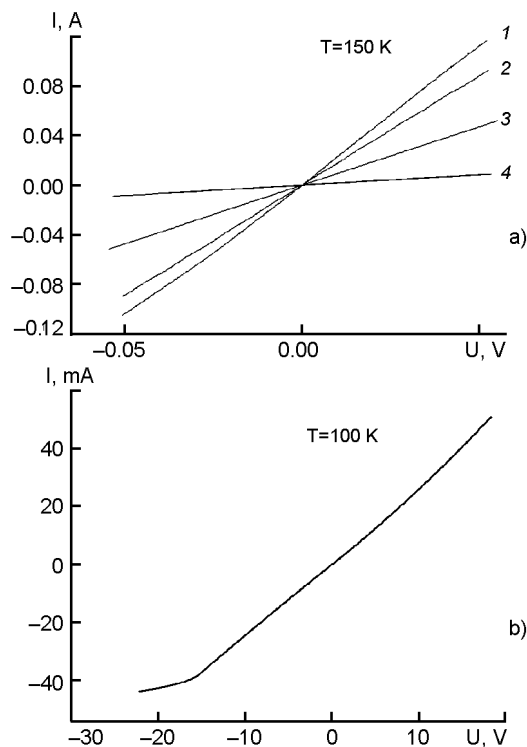

Fig. 5. The current-voltage characteristics of heterojunctions CdHgCrSe/MnHgTe.

As a result of indirect s–d exchange interaction the conduction band of the ferromagnetic semiconductor splits into two subbands due to the removal of degeneracy in the system of s-electrons. The growth of the spontaneous magnetization in the ferromagnetic temperature region is accompanied by the growth of the spin splitting of these subbands with the spin "up" (lower subband) and "down" (upper subband), thereby providing a variety of energy barriers for the tunneling electrons with different spin orientations. The magnitude of this splitting is determined by the following expression [19]:

$$\Delta_{sd} = \pm 1 / 2 S J_{nd} M(T) / M(0), \qquad (3)$$

where S — spin of the magnetic ion, J_{nd} — the value of s–d exchange integral, M — saturation magnetization at the current temperature and at $T = 0$ K. In the ferromagnetic semiconductor the conductivity is mainly occurs in the lower subband, which brings together all of the impurity levels. When applying a negative bias voltage in accordance with the magnetization of ferromagnetic semiconductor, tunneling of the spin-oriented electrons occurs in the upper subband. At a certain bias the conductance

will tend to the saturation due to a decrease of the charge carriers mobility because of the increase of their scattering by magnons. This process is manifested in the form of the observed non-linear part of the current-voltage characteristic (Fig. 5b).

5. Conclusions

In this paper we studied the structural and magnetoresonance properties of a quaternary solid solution CdHgCrSe. The temperature dependence of the EPR spectra parameters indicate an establishment of ferromagnetic ordering in this compound at the temperature $T_C = 124$ K. This semiconductor has been used as an element of new heteropair HgCdCrSe/HgMnTe. The obtained temperature dependencies of electrical resistivity and magnetoresistance are typical for DMSs in the region of their transition from the paramagnetic to the ferromagnetic phase. In the ferromagnetic temperature range the non-linear part of the current-voltage characteristic with a negative bias voltage applied to the heterojunction has been experimentally observed that may indicate the presence of spin-polarized current in the given heterostructure.

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HgCrCdSe як елемент нової гетероструктури HgCrCdSe/HgMnTe

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З застосуванням метода ЕПР показано, що введення у твердий розчин $Cr_xHg_{1-x}Se$ четвертого компонента у вигляді кадмію (Cd) дозволяє підвищити температуру переходу даної сполуки у фазу магнітного упорядкування. Електроопір, магнітоопір та вольт-амперні характеристики нових гетеропереходів HgCdCrSe/HgMnTe виміряно в залежності від температури та зовнішнього магнітного поля. Поява нелінійної області на зворотній гілці експериментальних вольт-амперних характеристик при температурах, нижчих за температуру магнітного упорядкування, може бути пояснено з точки зору наявності спін-поляризованого тока.