Quaternary semimagnetic $Hg_{1-x-y}Cd_xMn_ySe$ crystals for optoelectronic application

Yu.I. Mazur, G.G. Tarasov, E.V. Kuz'menko, A.E. Belyaev, W. Hoerstel*,
W. Kraak*, W.T. Masselink

 Institute of Semiconductor Physics, NAS of Ukraine,
45, prospect Nauki, 03650 Kyiv, Ukraine
**Humboldt-Universitat zu Berlin, Dept. of Physics, Invalidenstrasse*
110, D-10115 Berlin, Germany

Abstract. The peculiarities of the exchange interaction between the carrier spin and localized spin moments of magnetic ions in the close vicinity of semimetal-semiconductor transition have been studied on the example of semimagnetic quaternary solid solution $Hg_{1-x-y}Cd_xMn_ySe$ ($x = 0.10, y = 0.02$, $E_g = 45$ meV) by means of Shubnikov-de Haas oscillations. It is shown that due to the *s-p* hybridization of electron wavefunctions the exchange constant α turns out in a complicated function $\tilde{\alpha}$ of energy gap E_g and electron concentration which changes from positive $\tilde{\alpha}N_0 = 0.15$ eV to negative $\tilde{\alpha}N_0 = -0.28$ eV values which are typically accepted for wide gap semimagnetic semiconductors.

Keywords: semimagnetic semiconductor, constant of exchange interaction, semimetal-semiconductor transition.

1. Introduction

It has been demonstrated that «spin» doping of semiconductors leads to a drastical changes in their optical and transport properties [1, 2]. These changes originate from the exchange interaction between the conduction electron spin and the localized magnetic moments of magnetic ions, typically, Mn ions. Exchange interaction modifies the energy band structure and introduces a striking differences between the quantum transport phenomena observed in diluted magnetic and nonmagnetic semiconductors [3, 4]. Usually in nonmagnetic semiconductors the amplitude of a given magnetoresistance maximum decreases monotonically with temperature increase, whereas the oscillatory behavior of magnetoresistance can be observed in semimagnetic materials.

The most suitable conditions for the Shubnikov-de Haas (SdH) oscillations observation are met in narrow-gap materials due to a small value of a free carrier effective mass. Mixed $A^{II}B^{VI}$ compounds $Hg_{1-x}Mn_xTe$ and $Hg_{1-x}Mn_xSe$ allow gradual transition from the zero-gap (semimetals HgTe, HgSe) to the open-gap (semiconductor) state by increasing the manganese content. However, when the semimetal-semiconductor transition has happened the manganese content occurs high enough to generate a numerous magnetic clusters which strongly affect the spin-dependent phenomena [5, 6]. In order to avoid this obscuring influence the energy gap in HgTe or HgSe can be preferably opened by adding the cadmium with the subsequent introduction of magnetic ions in mixed $Hg_{1-x}Cd_xTe$ or $Hg_{1-x}Cd_xSe$ crystals. Quaternary compounds $Hg_{1-x-y}Cd_xMn_yTe$ and $Hg_{1-x-y}Cd_xMn_ySe$ demonstrate improved optical and transport characteristics in comparison with those in ternary $Hg_{1-x}Cd_xTe$ and $Hg_{1-x}Cd_xTe$ when their composition is matched properly [7–9]. In this paper the $Hg_{1-x-y}Cd_xMn_ySe$ single crystals with small energy gap E_g, $E_g \sim 50$ meV, are studied. The manganese concentration is kept to be a comparatively low ($y \sim 0.02$). In this case the Brillouin function describes the crystal magnetization reasonably well.

The investigation is aimed to study the peculiarities of exchange interaction in the close vicinity of semimetal-semiconductor transition. This interaction is typically described by two basic exchange constants, α and β, which present *s-d* and *p-d* exchange integrals, respectively [10, 11]. The sign of parameter α, determined experimentally, is negative, whereas β is of a positive value and differs from α approximately by factor 2 in $Hg_{1-x}Mn_xTe$ [12]. For the wide gap diluted semimagnetics the values of α and

β are reproducible with a great accuracy, whereas for narrow-gap semiconductors these values, determined by different authors, e.g., for Hg$_{1-x}$Mn$_x$Se, demonstrate a considerable scattering [4, 13-15]. This latter mirrors the complicated nature of the band structure in narrow-gap compounds, where both spin and orbital quantization should be taken into account simultaneously. Below we present the data which make it questionable even the separate usage of α and β for the description of the SdH oscillations behavior in narrow-gap Hg$_{1-x-y}$Cd$_x$Mn$_y$Se, at least.

2. Experiment

The monocrystalline Hg$_{1-x-y}$Cd$_x$Mn$_y$Se samples were grown by the modified Bridgman method. The composition of the samples was determined by microprobe analysis. For the SdH measurements were used the homogeneous samples only. According to the results of microprobe analysis the Mn distribution over the sample area differs by less than 0.1 at %. The measurements were performed on the samples having the composition $x = 0.10$, $y = 0.02$. As-grown crystals had the electron concentration $n = (3-5) \cdot 10^{17}$ cm^{-3}. In order to change the free-carrier concentration the samples were annealed in the vapor of the components. Due to heat treatment the electron concentration was varied in the range $(0.4-20) \cdot 10^{17}$ cm^{-3}. The magnetoresistance has been measured in magnetic field up to $15T$ and temperature range 1.5–80 K. The samples used for SdH measurements were sliced into a rectangular Hall-bars with dimensions of $9 \times 1.5 \times 1.2$ mm^3. Ohmic contacts were obtained by soldering with 10% tin-indium. The Hall coefficient values did not depend essentially on temperature due to degeneration of electron gas. The value of electron mobility μ_n at $T = 4.2$ K approaches the value $3 \cdot 10^5$ cm^{2}V^{-1}s^{-1} for $n = 1.6 \cdot 10^{17}$ cm^{-3}, being to best of our knowledge, the same order as that in the state-of-the art Hg$_{1-x}$Cd$_x$Se. The parameters of the samples used in our measurements are given in Table 1.

Here the electron concentrations n were derived from the period of SdH oscillations. They coincide within 2% accuracy with those found from Hall measurements. The E_g value was determined from the optical studies in narrow-gap Hg$_{1-x-y}$Cd$_x$Mn$_y$Se [9]. Parameters m_n and m_e are the electron mass at bottom of conduction band and the mass of free electron, respectively. The effective mass m_F and the gyromagnetic factor g_0 at Fermi energy were calculated following [16] in the Kane approximation with account of the contribution of higher energy bands. The Luttinger parameters, the Kane momentum matrix element P, and the spin-orbit splitting Δ were those for Hg$_{1-x}$Mn$_x$Se [13]: $g_1 = 3.0$, $g_2 = 0.5$, $g_3 = -0.17$, $k = -1.5$, $L = 0$; $P = 7.2 \cdot 10^{-8}$ eV cm, and $\Delta = 0.39$ eV.

The composition used has been chosen due to following experimental fact. It has been established [17] that in Hg$_{1-x}$Cd$_x$Se with $x = 0.15$ (semiconductor state) unusual strong SdH oscillations develop and their positions in transverse (ρ_{xx}) and longitudinal (ρ_{zz}) magnetoresistance are not shifted by phase. The SdH patterns demonstrate well resolved spin split structure. Following these find-

ings we take the composition $x = 0.1$ and $y = 0.02$, because at zero magnetic field B the energy gap in Hg$_{1-x-y}$Cd$_x$Mn$_y$Se is opened in the same manner as in Hg$_{1-x'}$Cd$_{x'}$Se with effective concentration $x' = x + 2.52y$ (see Ref. [9]). With such adjusted composition we believe to meet the most favorable conditions for observation of exchange interaction effect on the SdH spin-related structure in Hg$_{1-x-y}$Cd$_x$Mn$_y$Se.

3. Results and discussion

A. Experimental results

Fig. 1 depicts the SdH patterns for the ρ_{xx} and ρ_{zz} magnetoresistance in sample 1 at different temperatures. Due to low electron concentration the B_0^- maximum for ρ_{xx} is observed at $B \approx 6$ T. Below it, the spin split maxima $B_0^{\pm}$ are well resolved at low temperature ($T = 1.5$ K) with subsequent smearing them at temperature increase. Comparison of this dependence with that presented in Ref. [16] for non-magnetic Hg$_{1-x}$Cd$_x$Se of proper cadmium content shows a remarkable similarity. The SdH dependence for ρ_{zz} reproduces all the maxima of ρ_{xx} without a visible phase shift at low temperature. It is clearly seen also that B_N^+ maxima ($N \geq 1$) are stronger than those of B_N^- in the ρ_{xx} dependences, whereas B_N^- dominate B_N^+ peaks in the ρ_{zz} dependences. With temperature elevated the B_0^- maximum shifts towards higher magnetic field as in Fig.2. The spin splitting δ_N, $\delta_N = B_N^+ - B_N^-$, decreases for all Landau numbers $N \geq 1$. The linear dependences of the integer versus reciprocal magnetic fields for the ρ_{xx} and ρ_{zz} spin split components, shown in Fig. 3, allow to determine precisely the period of SdH oscillations and to calculate the electron concentration n (see Table 1).

Fig. 4 presents the temperature-induced evolution of the SdH dependence for ρ_{xx} in the sample 2. In view of higher electron concentration the B_0^- maximum occurs at $B \approx 12$ T. The spin splitting is obviously seen for all $N \leq 7$ peaks at $T = 1.5$ K. With temperature increase δ_N weakly reduces as shown by insert in Fig. 4. The temperature-induced shift of B_0^- maximum is plotted in Fig. 5. The peak motion toward higher magnetic field becomes more energetic when temperature passes $T \approx 15$ K.

Fig. 6 shows the modification of the ρ_{xx} dependence in sample 4 with temperature growth. For the high electron concentration the B_0^- maximum is far beyond our experimental facilities. We surely observe the magnetoresistance peaks with $N \geq 6$. These peaks being initially non-split at low temperature reveal a well resolved spin structure at higher temperature. The value of δ_N grows with temperature increasing, as shown by divergent dashed lines in Fig. 7.

Fig. 8 summarizes our findings on the effective parameter $\tilde{\alpha}$ variation with the electron concentration n in the vicinity of semimetal-semiconductor transition in the Hg$_{1-x-y}$Cd$_x$Mn$_y$Se single crystals. This parameter is introduced to describe the exchange interaction effect in semimagnetic narrow-gap compounds.

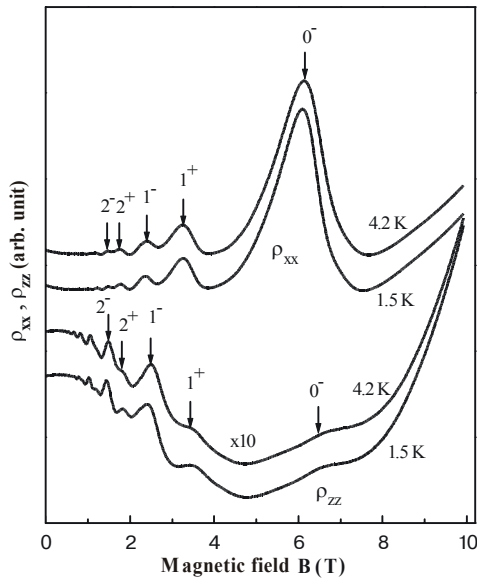Fig.1. SdH oscillations patterns for Hg$_{1-x-y}$Cd$_x$Mn$_y$Se single crystal in transverse (ρ_{xx}) and longitudinal (ρ_{zz}) configurations. The oscillation peaks are labeled by the Landau number N and the sign for the spin state. Sample 1: $x = 010$, $y = 0.02$; $n = 4.4 \cdot 10^{16}$ cm^{-3}

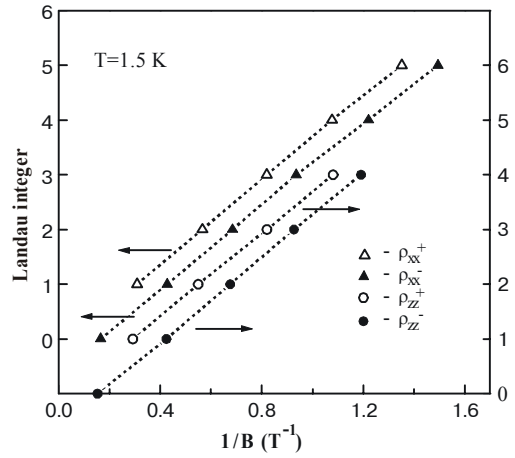Fig.3. Plot of the Landau integer versus reciprocal magnetic fields: triangles stand for the ρ_{xx} magnetoresistance, circles are for the ρ_{xx} oscillations. Sample 1, $T = 1.5$ K
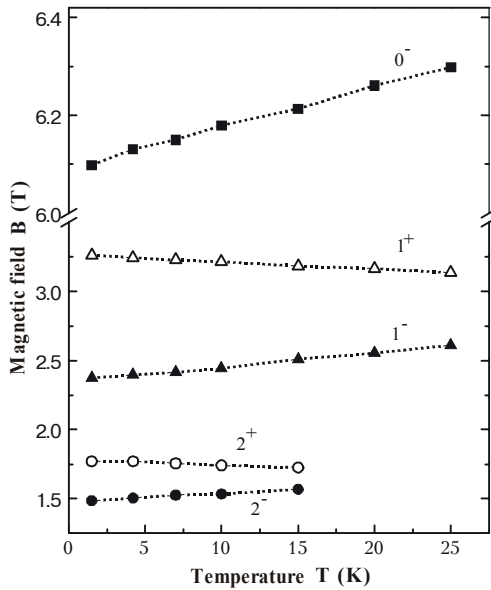

Fig.2. Temperature-induced shift of the spin split components for the first SdH maxima of ρ_{xx} magnetoresistance ($N = 0, 1, 2$) in sample 1.
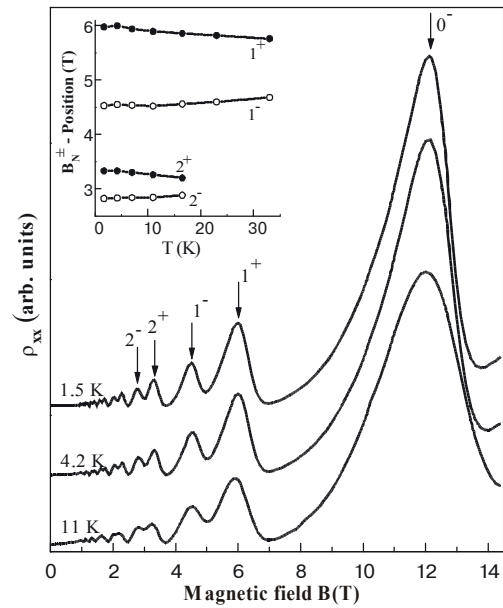

Fig.4. A temperature variation of the SdH patterns in ρ_{xx} configuration for Hg$_{1-x-y}$Cd$_x$Mn$_y$Se, Sample 2. The peak shift caused by the change of temperature for $N = 1, 2$ is plotted by insert. Sample 2: $x = 010$, $y = 0.02$; $n = 1.08 \cdot 10^{17}$ cm^{-3}

B. Discussion

In order to start it should be mentioned that both the B_0^- speak position and the spin splitting of B_N peaks of magnetoresistance are defined to the great extent by the value of gyromagnetic factor g_0 at the Fermi level. Being negative at $B = 0$, g_0 decreases by module, as can be seen from Table 1, with n increase following approximately the dependence
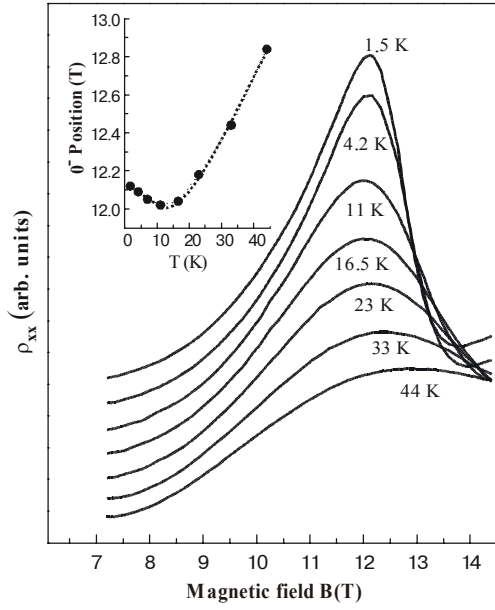
Fig.5. Plot of B_0^- maximum motion in the ρ_{xx} dependence on temperature in Sample 2. Nonmonotonic shift of B_0^- is shown by insert.
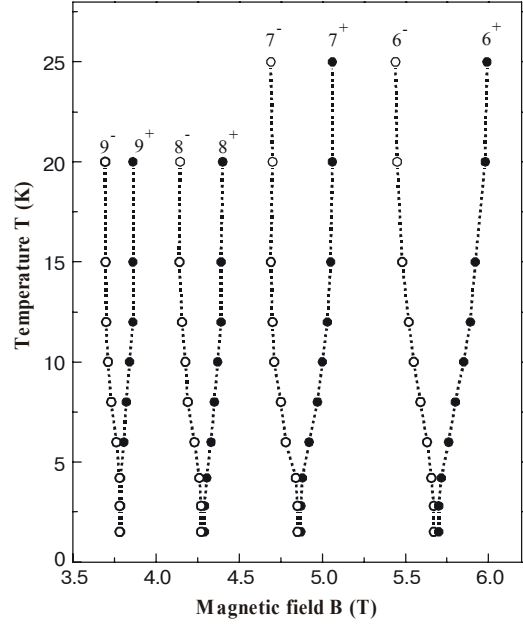
Fig.7. Thermally induced divergence of spin-splitted maxima in ρ_{xx} configuration in Sample 4.
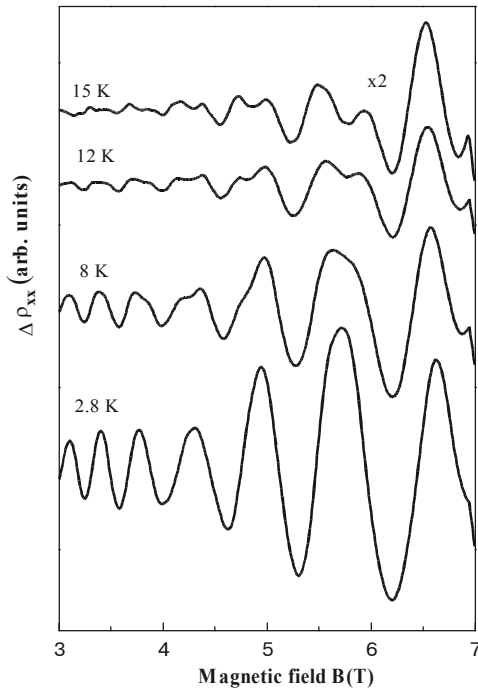
Fig.6. Temperature-induced transformation of the SdH pattern in the ρ_{xx} configuration for Hg$_{1-x-y}$Cd$_x$Mn$_y$Se, Sample 4: $x = 010$, $y = 0.02$; $n = 1.21 \cdot 10^{18}$ cm^{-3} . The spin related structure develops at higher temperatures.
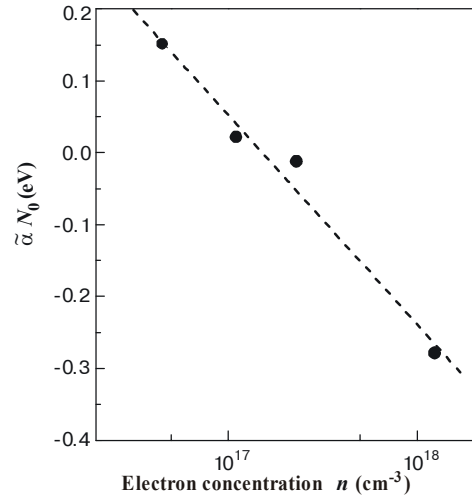
Fig.8. Plot of the $\tilde{\alpha}N_0$ function on the electron concentration n. Compilation of the data for Samples 1–4. Line is for eyeguidance.

which has been derived from the Kane theory, when the influence of remote energy bands was neglected [18]. In magnetic field the g_0-factor is contributed by exchange interaction and becomes renormalized to [12]

$$g^* = g_0 + N_y \alpha \langle S_z \rangle / \mu_B H \qquad (2)$$

for the Γ_6 band and to

$$g^* = g_0 + N_y \beta \langle S_z \rangle / 3\mu_B H \qquad (3)$$

for the Γ_6 band in the semimagnetic semiconductors.

In equations (2) and (3) $N_y = yN_0$, where N_0 is the number of unit cells per unit volume, y is the Mn molar

$$g_0 = 2\left\{1 + \left(1 - \frac{m_e}{m_F}\right)\frac{\Delta}{3E_F + 3E_g + 2\Delta}\right\} \qquad (1)$$

fraction, and μ_B is the Bohr magneton. The thermal average $\langle S_z \rangle$ of the spin operator $\hat{S}$ projection z is determined by the Brillouin function [1]

$$\langle S_z \rangle = -S_0 B(\xi) \tag{4}$$

and

$$B(\xi) = \frac{2S+1}{2S} \coth\left(\frac{2S+1}{2S}\xi\right) - \frac{1}{2S}\coth\left(\frac{1}{2S}\xi\right)$$

$$\xi = g_{Mn}\mu_B SH / k_B (T + T_0)$$

Here $g_{Mn} = 2$ is the gyromagnetic factor of the Mn ion, $S = 5/2$. Empirical parameters S_0 and T_0 take into account the existence of magnetic clusters and the antiferromagnetic interaction between the manganese ions, respectively.

The sign of exchange parameter α is negative and its contribution to g^* (equation 2) is positive. Therefore the exchange induced contribution to electron g-factor has to reduce the latter by module in open-gap semiconductor (Γ_6 energy band). For zero-gap semimagnetics (Γ_8 band) the exchange contribution is of the same sign as the g_0-factor ($\beta > 0$) and, hence, g^* has to be enhanced by module. Following these arguments, one can expect in open-gap materials that the exchange contribution, being a sizable fraction of the total spin splitting for the conduction electrons, will favor a significant reduction of the electron spin splitting at very low lattice temperature. With temperature increasing this contribution rapidly decay due to reduction of $\langle S_z \rangle$ (Eq. 4) and the total spin splitting has to be increased. Contrary, in zero-gap material the temperature increase produces a reduction of the spin splitting by the same reason.

Let us go to discussion of temperature behavior of SdH oscillations in sample 1. The position of B_0^- maximum can be determined from [19]

$$B_0^- = \frac{\hbar c}{e}\left(\frac{2\pi^4 n^2}{|\gamma|}\right)^{1/3}, \tag{5}$$

where $\gamma = \frac{m_F}{2m_e}g^*$. The peak position shift observed experimentally (Fig. 2) can be connected immediately with the change of the parameter $|\gamma|$ value since the electron concentration is suggested to be constant at temperature variation due to degeneration of electron gas. Hence, one concludes from the experimental data that as far as B_0^- peak moves towards higher magnetic field with temperature increase the $|\gamma|$ value has to decrease. For the multiplier $(m_F / 2m_e)$ definitely grows with the temperature elevating the $|\gamma|$ reduction can originate from the $|g^*|$ reduction only. Because of the open-gap nature of the Hg$_{1-x-y}$Cd$_x$Mn$_y$Se crystal under investigation here the $|g^*|$ temperature behavior is governed by Eq. 2. The first term in the right part of this equation, g_0, can be reduced by module in principle with temperature increase due to en-

largement of E_g and E_F (see Eq. 1). It was determined from the earlier optical measurements (see Ref. [9]) that in the Hg$_{1-x-y}$Cd$_x$Mn$_y$Se crystals of similar composition dE_g / dT equals to 0.54 meV/K. When this temperature-induced «opening» the energy gap is taken into account its contribution to the total B_0^- peak shift gains approximately 12% of that observed experimentally only. Therefore the contribution of exchange related term in equation 2 should be dominating. However, if the α sign is assumed to be negative (the case of open-gap semimag-netics) one meets a severe contradiction with experiment: either an unreasonable high exchange contribution at $y \approx 0.02$ should be admitted in order to make the sum g^* positive when α is negative, or the α sign is to be accepted positive.

In our earlier SdH experiment [20] with the Hg$_{1-x-y}$Cd$_x$Mn$_y$Se crystals of similar composition it was established that in order to agree the SdH data with theory, operating with two independent α and β, the module of α must be reduced by factor 1.5–2. Unfortunately, in those experiments the B_0^- maximum was not reached and the conclusion was derived from the temperature dependence of the spin splitting of the SdH maxima. This dependence was found to be weak and did not allow the more definite assignment. Nevertheless, it was supposed that such α reduction could arise from the admixture of p-like wavefunctions to the s-like wave-functions of the conduction band electrons. In this case the exchange contribution has to be taken into account by substitution of $(F_1\alpha + F_2\beta)$ instead of α into equation 2. The F_1 and F_2 functions depend on the Kane's coefficients which include the contribution of remote energy bands [21].

The SdH results presented here are much more indicative, because the B_0^- maximum is easily seen and it is possible to trace its movement with temperature variation. Using the data from Table 1 and the position of B_0^- peak the $|g^*|$ value has been derived and is found to be $|g^*| = 64$, whereas $|g_0| = 47.5$ (see Table 1). The number obtained positively states that the exchange contribution to $|g^*|$ is amplifying one, i.e., the exchange parameter α occurs positive by sign even for the Γ_6 energy band. This crucial result completely changes the physical picture and proves that in the close vicinity of semimetal-semiconductor transition ($E_g > 0$) the exchange contribution cannot be described simply in terms of two independent exchange constants, as it was done in the approximation of molecular field. Nevertheless, this contribution stays to be proportional to $\langle S_z \rangle$ and manganese concentration N_y. Therefore, we assume that equation 2 holds its shape, but the physical meaning of parameter α is different: α cannot be considered as a constant but is the function of E_g in the range of small (positive) E_g. Since we will imply further an effective parameter $\widetilde{\alpha}$ instead of α when appeal to equation 2. At low temperature the $\langle S_z \rangle$ function tends its saturation value S_0, which typically is less than 5/2 due to clustering effect. In this temperature range the g^* value is to be the largest. When temperature is lifted up the $\langle S_z \rangle$ value reduces by module and the rate of reduction is defined by effective temperature T_0. Following our experimental results the T_0 value should be

large enough, $T_0 \approx 8$ K, in the sample 1. Under these conditions the B_0^- peak motion with temperature increasing towards higher magnetic field, seen in Fig. 2, becomes well motivated.

Table 1

Hg$_{1-x-y}$Cd$_x$Mn$_y$Se single crystal $x = 010, y = 0.02, E_g = 45$ meV, $T = 4.2$K					
Sample label	$n \cdot 10^{17}$, cm^{-3}	$\dfrac{m_n}{m_e} \cdot 10^3$	$\dfrac{m_F}{m_e} \cdot 10^2$	g_0	E_F, meV
1	0.44	4.7	1.48	-47	44
2	1.08	4.7	1.94	-36	68
3	2.33	4.7	2.46	-28	93
4	12.1	4.7	4.13	-15	183

The temperature variation of spin splitting for the SdH maxima is shown in Fig. 2. The peak positions are determined by [19]

$$B_N^\pm = \frac{\hbar c}{e}\left(\sqrt{2}\pi^2 n\right)^{2/3} \left\{ \sum_{k \geq 0, \mp |\gamma|}^{N} \left(\sqrt{k} + \sqrt{k \pm |\gamma|} \right) \right\}^{2/3} , N \geq 1 \quad (6)$$

Here the thermal smearing of Fermi energy is not taken into account and equation 6 is, strictly speaking, valid only at very low temperature. If $|\gamma| = 0$ or $|\gamma| \ll 1$ the SdH maxima can not be resolved practically. In the case of sample 1 the $|\gamma|$ value has been calculated from the ratio B_1^+ / B_1^- and occurs to be large enough, $|\gamma| = 0.46$, resulting in a well resolved spin splitting at low temperature. The $|g^*|$ value determined from the spin splitting of the $N = 1$ SdH peak is found to be equal $|g^*| = 62.3$, which reproduces the value derived from the B_0^- position within the 2.5 % accuracy. With the temperature increase the $|\gamma|$ value, found from the temperature variation of δ_N, reduces and the spin splitting δ_N decreases as shown in Fig. 2. Hence, the temperature behavior of the δ_N value is also consistent with the general picture enlightened above. It should be mentioned here that the spin splitting δ_N can be effectively reduced also due to thermally-induced broadening of the SdH peaks, typically as in the case of nonmagnetic semiconductors [17]. Therefore, the smearing of spin split structure at higher temperature is not so indicative as the motion of the B_0^- peak. Indeed, the consideration which takes carefully into account incomplete degeneration of electron gas as well as the broadening of Landau levels due to collisions (the Dingle temperature), gives the corrections to $|\gamma|$ facilitating the motion of B_0^- maximum towards the lower magnetic field, in the direction upright opposite to that observed experimentally.

The electron concentration n in sample 2 differs by 2.5 factor against that in sample 1. Fig. 4 reproduces the SdH oscillations patterns in this sample at three different temperatures. A well-pronounced spin splitting accompanies each N-th SdH maximum at low temperature with subsequent smearing the spin structure at higher temperature ($T \sim$ 40 K). Basing on our findings for sample 1 we start the analysis from the temperature behavior of B_0^- maximum located at $B \sim 12$ T. The non-monotonic shift of the B_0^- peak with temperature is mirrored by the insert in Fig. 5. Up to temperature $T \approx 15$ K the peak moves slightly towards lower magnetic field, but then it changes the direction of motion and goes quickly up to higher magnetic field. Using the B_0^- peak position, as well as, the spin splitting δ_1 at $T = 1.5$ K (Fig. 4, insert) the $|g^*|$ value was derived. It occurs to be a surprisingly close the $|g_0|$ value in sample 2: $|g^*| = 37$ against $|g_0| = 36$. According to equation 2 this indicates either no contribution of the exchange interaction in sample 2 or a very small negative contri-bution to g^* if the latter exists. One could even suspect whether the manganese ions present in this sample, but our earlier studies by means of far-infrared phonon spectroscopy (reflection, transmission [9]), as well as the data of microprobe analysis surely verify the manganese presence of a proper concentration. Whence we conclude that the exchange contribution in the range of small E_g of semimagnetics becomes also strongly dependent on electron concentration being nearly suppressed at $n \approx 1 \cdot 10^{17}$ cm^{-3}. If this statement is true the low-temperature behavior of the B_0^- peak and the spin- splitting of the SdH maxima can be reasonably explained. Indeed, due to thermal smearing of the Fermi edge the B_0^- peak position is determined by equation 5 with $|\gamma|$ changed by $|\tilde{\gamma}|$:

$$|\tilde{\gamma}| = \left(\sqrt{|\gamma|} + 0.535 \sqrt{k_B T m_F c / \hbar e B_0^-} \right)^2 . \quad (7)$$

As a result, the B_0^- position is to be normally shifted to the side of lower magnetic field with temperature increase. This fact is mirrored by the lowering part of curve in Fig. 5 (insert). The similar corrections can be introduced in equation 6 also, but the thermal shift of $B_N^\pm$ maxima will be less pronounced in comparison to that for the B_0^- peak. The spin splitting is also smeared with temperature growth: besides diminishing the oscillations amplitudes, the reconciliation of the spin split doublet components and their broadening lead to complete disappearance of the spin related structure of the SdH maxima. Hence, in the case of sample 2 we observed the behavior typical for nonmagnetic semiconductors when temperature is below 15 K.

The further shift of the B_0^- maximum towards higher magnetic field after the temperature passes $T = 15$ K seems to be of an activation nature. Indeed, when magnetic field is strong enough ($B \sim 12$ T), the localization of electrons at the fluctuations of potential is of high probability. At the B strength which corresponds to the maxima of Hall coefficient oscillations, the localization can be of a resonant character [22] and the electrons which possess a low kinetic energy (slightly above the Landau sub-bands bottom) become localized. The ratio of the localized electrons number to their total quantity is not very small. The resonant localization can be a reason of unusually large amplitudes of the Hall oscillations and those for the ρ_{xx}. In our case such sort of localization could develop at low

temperature ($T \leq 15$ K). The temperature increased above 15 K releases the trapped electrons, effectively affecting the electron concentration n. This latter results in the B_0^- peak shift towards higher magnetic field (see equation 5) without a noticeable change of the $|\gamma|$ value. Therefore, the spin splitting of the SdH maxima does not react immediately on the more energetic B_0^- peak shift as shown in Fig. 4 (insert).

The further n increase by factor 2.2 does not change basically the SdH oscillations behavior against that observed in sample 2. In sample 3 the $B_N^\pm$ doublet was surely detected at $B \sim 9$ T while the B_0^- peak was beyond our facilities. The $|g^*|$ value is derived to be $|g^*| = 26$ from the δ_I spin splitting , whereas $|g_0| = 28$ in sample 3. It shows that the exchange interaction is not yet actual in the case. Whenever the electron concentration was increased up to $1.21 \cdot 10^{18}$ cm^{-3} the SdH oscillations behavior becomes distinctly different. Fig. 6 shows the ρ_{xx} variation versus magnetic field up to $B = 7$ T, when the most prominent changes of the SdH peaks develop with temperature increase. The low temperature patterns do not reveal any spin splitting. When temperature was elevated, the well resolved structure of the B_N peaks with $N \geq 6$ was detected. The splitting grows with temperature and is saturated when temperature reaches 15 K for the most of the SdH maxima (Fig. 7). Further temperature increasing leads to a significant broadening of the split components and the thermally induced structure smearing. In terms of the $|g^*|$ factor the latter means that $|g^*| \approx 0$ (δ_N is beyond the resolution for $N \geq 6$) at low temperature ($T = 1.5$ K) and reaches its maximal value $|g^*| = 18$, derived from the δ_N splitting at T = 15 K. This latter is found to be close to the $|g_0|$ value in sample 4, $|g_0| = 15$. Following the arguments presented above one can conclude that function $\tilde\alpha$, which introduces the exchange contribution in equation 2, takes now a negative value, normally for open-gap semimagnetics. At low temperature this contribution completely equalizes g_0 and the $|g^*|$ value turns out the zero leading to a disappearance of the SdH peak spin splitting. With temperature increase the exchange contribution decays due to effective $\langle S_z \rangle$ reduction and one observe normally spin split maxima.

Taking into account our findings for all the samples investigated the function $\tilde\alpha$ versus the electron concentration n can be derived. Let us assume that the S_0 and T_0 parameters determining the $\langle S_z \rangle$ dependence are of the same value for all samples. The T_0 value derived from the temperature dependence of δ_N is taken to be 8 K. The value of clustering parameter S_0 was calculated taking into account the probability of various magnetic clusters realization when a stochastic distribution of magnetic ions over the crystal lattice is assumed [5,6]. For $y = 0.02$ the S_0 value is of 2.1. Under these assumptions the $\tilde\alpha(n)$ dependence is that shown in Fig. 8. Its magnitude varies from positive ($\tilde\alpha N_0 = 0.15$ eV) to negative ($\tilde\alpha N_0 = -0.28$ eV) one when the electron concentration n is within the range $4.4 \cdot 10^{16} \leq n \leq 1.21 \cdot 10^{18}$.

Until we deal with the peculiarities of exchange interaction in narrow-gap semimagnetic semiconductor Hg$_{1-x-y}$Cd$_x$Mn$_y$Se which reveal themselves in the tempera-turedependent behavior of the SdH oscillations. These peculiarities are connected with the complicated nature of the energy band structure, when both spin and orbital quantization have to be taken into account. New proofs of such hybridization arise from the comparison of the ρ_{xx} and ρ_{zz} magnetoresistances shown in Fig. 1. The SdH maxima coincide in the ρ_{xx} and ρ_{zz} dependences within the experimental accuracy for all Landau numbers $N \geq 0$. Typically the spin split structure is more distinct in the ρ_{xx} than in the ρ_{zz} dependence reflecting the difference in the nature of transverse and longitudinal magnetoresistances [23, 24]. It follows from the theory of longitudinal magnetoresistance [24] that at $T = 0$ and without damping to be taken into account both the $B_N^\pm$ peaks have to be of equal strength. But, it was proved experimentally that the B_N^+ peaks are either of much smaller intensity if observed or are absent completely [25–27]. Efros [28] has shown that when inequality $k_B T_D \langle\langle k_B T \langle\langle \mu_B H$ takes place (T_D is the Dingle temperature, which determines the Landau level broadening due to collisions), the maxima of the ρ_{xx} and ρ_{zz} oscillations coincide. Nevertheless, when the spin-flip transitions are of a negligible probability the B_0^- maximum in the ρ_{zz} dependence is forbidden. Suizu and Narita [29] have found that if only a simple s-like wavefunctions are considered for the conduction band both the B_N^+ and B_0^- peaks are prohibited. However, if one takes into account the p-like contribution of the conduction band wavefunction these peaks should be allowed again. Besides, the direct calculation of the non thermal broadening contribution to the Landau levels also give the amplitudes of the B_N^+ peaks in the ρ_{zz} dependence much smaller, than those for the B_N^- [30]. Following our data we conclude that the clear manifestation of the B_0^- peak gives evidence for the spin-flip transitions in the narrow-gap Hg$_{1-x-y}$Cd$_x$Mn$_y$Se. These transitions become very intensive when $E_g \to 0$, and the electron wavefunction is hybridized from the s-like and p-like states. This conclusion is in line also with our results for quaternary Hg$_{1-x-y}$Cd$_x$Mn$_y$Te with $E_g \sim 120$ meV where the spin-flip stransitions where detected both in photoluminescence and in the SdH oscillations [31]. The well resolved spin split structure in the ρ_{zz} oscillations and the absence of a visible shift between the ρ_{xx} and ρ_{zz} maxima prove also a good quality of the samples under investigation. Otherwise, when T_D is high enough, the spin related structure has to be smeared.

Conclusions

The studies of the SdH oscillations behavior in narrow-gap semimagnetic semiconductor Hg$_{1-x-y}$Cd$_x$Mn$_y$Se enlightened novel aspects of the exchange interaction between the conduction band electron spin and the localized spin moments of magnetic ions which reveal themselves in the close vicinity of the semimetal-semiconductor transition. It has been demonstrated that the hybridization of the s-like and p-like electron wavefunctions strongly modifies the exchange interaction related assignment of the electron states. As a result the simple description in terms of two independent exchange

constants α and β derived in the approximation of molecular field is not valid. The exchange parameter α transforms into a function $\tilde{\alpha}$ which is strongly dependent on the energy gap E_g and the electron concentration n. This function magnitude varies within wide limits: from positive $\tilde{\alpha}N_0 = 0.15$ eV to negative $\tilde{\alpha}N_0 = -0.28$ eV values which are typically accepted for wide gap semimagnetic semiconductors. However, it should be noticed that the normally negative value the function $\tilde{\alpha}$ accepts at a sufficiently high electron concentrations, $n \sim 10^{18}$ cm^{-3}, when the renormalization of the electron energy parameters is already of importance. The findings of this study explains to some extent a considerable scattering the values of the exchange parameters observed for the narrow-gap semimagnetic semiconductors which is typically ascribed to an incomplete knowledge of the crystal magnetization. Indeed, such parameters have to be introduced for a particular crystal of a given composition with account of a free carrier concentration. Besides novel interesting aspects of semimag-netics physics our results give also evidence for a high quality of a crystalline structure of quaternary semiconductors under investigation. The well resolved spin structure of the longitudinal and transverse magnetoresistances without of a visible shift between their SdH peaks proves, at least, a low T_D originated due to collisions. This finding is in line with the statement about improved crystalline structure of spin doped semiconductors, derived earlier from our optical measurements in quaternary semimagnetics [8, 9, 31]

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