Magnetization and Shubnikov-de Haas effect in diluted magnetic semiconductors $(Cd_{1-x-y}Zn_xMn_y)_3As_2$

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The results of the Shubnikov-de Haas (SdH) effect and magnetization of single crystals of $(Cd_{1-x-y}Zn_xMn_y)_3As_2$ (CZMA) are presented. Specimens with $x+y=0.3$ and $y=0.04$, 0.06 and 0.08, subjected to long-time thermal annealing, were studied. SdH effect was investigated in fields B up to 11 T in the temperature range $T=4.2$–20 K. Magnetization was measured at $B\leqslant 5.5$ T and 5 K$\leqslant T\leqslant$20 K. SdH oscillations were analyzed taking into account (i) the anomalous magnetic field dependence of the cyclotron mass observed recently in CZMA, and (ii) influence of the $s-d$ interaction to the effective g-factor (g_{eff}) of conduction electrons. The dependence of g_{eff} on B and T was introduced in the analysis of the SdH oscillations by fitting the magnetization data with the effective Brillouin function. Values of the exchange-independent part of g_{eff}, the exchange constant in the conduction band and the spin-dependent part of the Dingle temperature were determined. [S0021-8979(97)22408-4]

I. INTRODUCTION

Recently[1] the Shubnikov-de Haas (SdH) effect was studied in diluted II−V semiconductors $(Cd_{1-x-y}\, Zn_x\, Mn_y)_3As_2$ (CZMA) with $x+y=0.3$ and $y\leqslant 0.08$, and strong magnetic field dependence of the cyclotron mass according to the law $m_c(B)=m_c(0)+\alpha_c B$ [with $m_c(0)/m_0=0.028-0.042$ and $\alpha_c/m_0=0.008-0.012$ T^{-1}] was established in samples with $y=0.04$–0.08. The dependence of m_c on B has not been predicted theoretically or observed earlier in diluted magnetic semiconductors (DMS). Additionally, CZMA crystals demonstrate a set of unusual properties which in conventional DMS are less presented or not observed at all. Among them are the high-temperature spin-freezing effect, the anomalous magnetoresistance and the strong dependence of hopping conductivity on Mn concentration (see Refs. 1 and 2 and references therein). These phenomena were explained by taking into account the complex crystal structure of CZMA and the remanent disorder of its metallic sublattice.[2] This allows to suppose the same reason, remanent lattice disorder, to be responsible for the cyclotron mass anomaly observed in CZMA.[1] However, further investigations are required to support this conjecture.

Investigations of as-grown CZMA crystals with rather high spatial inhomogeneity of the electron concentration gave no evidence for the field or temperature dependence of the effective g-factor (g_{eff}) of the band electrons.[1] On the other hand, the sensitivity of g_{eff} to B or T is a characteristic feature of DMS and results from the $s-d$ exchange interaction between electrons and localized magnetic moments. Influence of the $s-d$ interaction to the band spectrum of DMS modifies the content of harmonics in the SdH oscillations in different field or temperature intervals. In a recent study[3] CZMA crystals were subjected to long-time thermal annealing to reduce the fluctuations of the carrier concentration. In the annealed crystals the dependence of m_c on B was observed along with presence of the first and the second harmonics in the spectrum of the SdH oscillations.[3] This demonstrates qualitatively the coexistence of the ''exchange-dependent'' g_{eff} and the anomalous magnetic field dependence of m_c in one and the same material. However, the analysis of the SdH effect performed in Ref. 3 is insufficient (see Section III) because no parameters characterizing the interaction of the electrons with localized magnetic moments have been determined.

Here we report the results of measurements of the SdH oscillations and magnetization on the annealed CZMA crystals. Investigations of both effects in the same specimens offer the possibility of elucidating more information about the $s-d$ interaction and spin-dependent scattering of conduction electrons, and complementing the earlier[1,3] analysis of the SdH effect in CZMA.

II. EXPERIMENTAL DETAILS

CZMA single crystals with $x+y=0.3$ and $y=0.04$, 0.06 and 0.08 were grown by a modified Bridgman method[1] and were subjected to thermal annealing in vacuum during four months at 100 °C. Magnetoresistance measurements were made in the temperature range from 4.2 to 20 K in the transverse magnetic field configuration ($B\perp j$) between 0−11 T by using the six-probe dc technique[4] and electrodes fixed by soldering. Magnetization measurements were performed between 5 and 20 K in magnetic fields up to 5.5 T by using a commercial SQUID magnetometer. The measurement is made by cooling the specimen from 300 K in zero field and recording the data by increasing step-wise the magnetizing field.

III. EXPERIMENTAL RESULTS

The SdH oscillations observed in the CZMA specimens with $y=0.04$, 0.06 and 0.08 are displayed in Fig. 1. The conductivity is presented in arbitrary units because only relative magnitudes of SdH amplitudes are significant. The SdH concentration determined from the frequency of the SdH maxima ($n_{\mathrm{SdH}}=2.4$, 2.1 and 2.8×10^{18} cm^{-3} for $y=0.04$, 0.06 and 0.08, respectively) is in excellent agreement with the Hall concentration n_R. Presence of the first and the sec-

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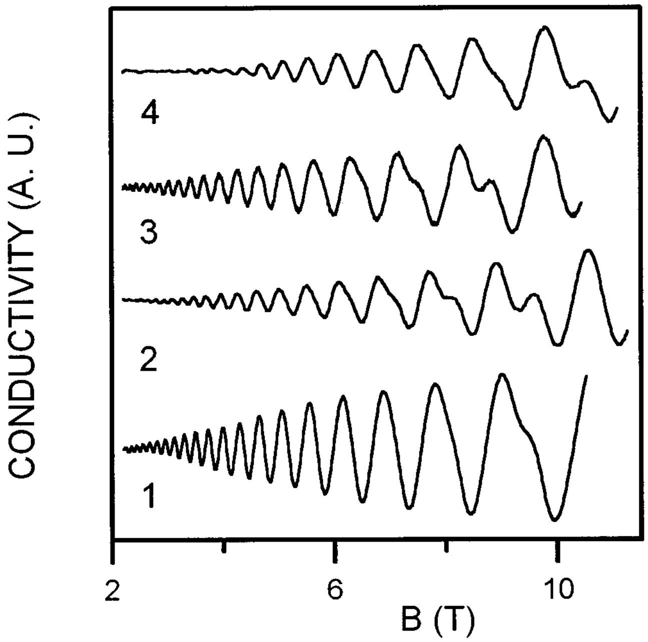

FIG. 1. SdH oscillations in the annealed CZMA crystals with $y=0.04$ at $T=4.2$ K (1) and 10.7 K (2), $y=0.06$ at $T=4.2$ K (3), and $y=0.08$ at $T=10.1$ K (4).

ond harmonics of the SdH oscillations is evident from Fig. 1 and is attributed to the exchange-dependent g_{eff}. The amplitudes of the first (A_1) and the second (A_2) harmonics were determined using a conventional method[5] (see Fig. 2).

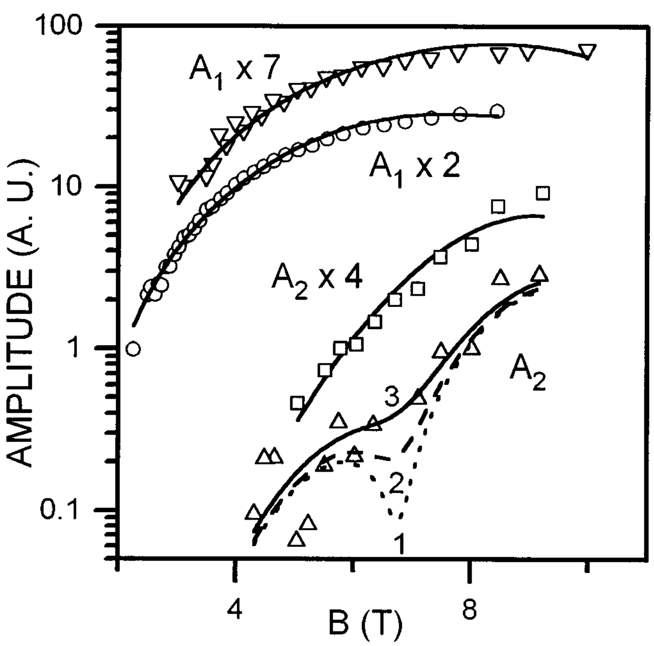

FIG. 2. Magnetic field dependence of the SdH harmonic amplitudes in a crystal with $y=0.04$ at $T=4.2$ K (○) and 7.0 K (▽) and in a crystal with $y=0.08$ at $T=4.2$ K (△) and 7.3 K (□). The solid lines represent the best fits of the experimental data to Eq. (1). Lines 1-3 are calculated with $\delta T_D=0.2$ K, 0.5 K and 0.9, K, respectively.

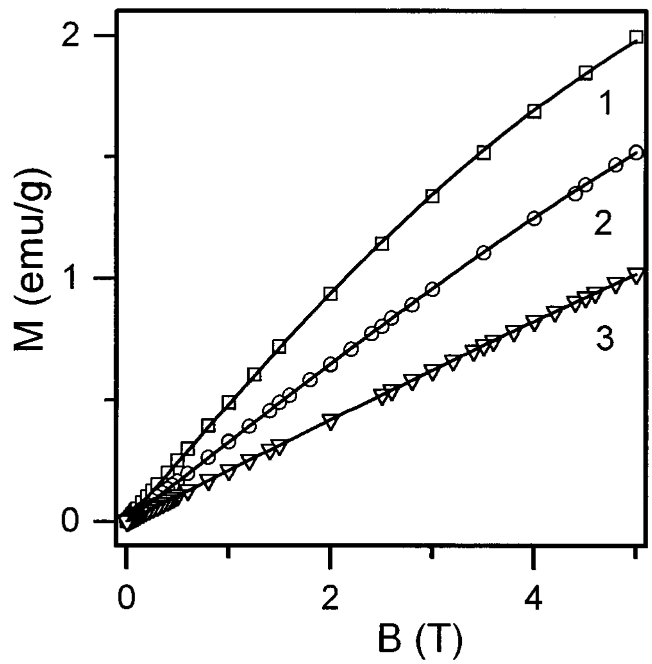

FIG. 3. Magnetic field dependence of the magnetization of the crystal with $y=0.04$ at 5 K (1), 10 K (2), and 20 K (3). The solid lines are the best fits of the experimental data with the values of $gS_0=2.15\pm0.02$, 2.45 ± 0.03 and 2.38 ± 0.07 and $T_0=5.5\pm0.1$ K, 7.7 ± 0.2 K and 6.8 ± 0.8 K for curves 1, 2 and 3, respectively.

In Ref. 3 the SdH oscillations measured on the same samples were analyzed in the field and temperature region where the effects connected with the $s-d$ interaction and spin-dependent scattering of conduction electrons can be neglected (i.e., omitting the exchange-dependent contribution to g_{eff}). The linear dependence of m_c on B with $m_c(0)/m_0=0.023-0.036$ and $\alpha_c/m_0=0.005-0.015$ T^{-1} was established by using two different methods. Values of the Dingle temperature, $T_D=18$–22 K, were found to be reduced compared with those determined for the as-grown crystals ($T_D=32$–39 K).[1]

With increasing temperature or magnetic field, A_2 approaches A_1 or even exceeds it (what is evident from Fig. 1) demonstrating influence of the s-d interaction to g_{eff}. This suggests that the effective g-factor of the band electrons is a function of temperature and magnetic field in the investigated samples. To take this into account (see Section IV), magnetization measurements have been performed. The dependence of the magnetization on the applied magnetic field is shown in Fig. 3 for one of the specimens.

IV. DISCUSSION

The amplitude of the rth harmonic of the SdH oscillations can be written as[5]

$$A_r=\frac{\sqrt{rB}}{\sinh(rz)}\exp\left(-\frac{rzT_D}{T}\right)F_r(T,B), \qquad (1)$$

TABLE I. Values of the exchange constant, αN_0, the exchange-independent part of the effective g-factor, g^*, and the spin-dependent part of the Dingle temperature, δT_D.

y	T (K)	$-\alpha N_0$ (meV)	$-g$	δT_D (K)
0.04	4.2	220±20	19±1	2.1±0.2
	7.0	250±20	17±1	0.9±0.1
	10.7	270±10	18±1	0
0.06	4.2	230±30	20±2	0.5±0.2
	9.1	270±40	20±3	0
0.08	4.2	230±20	18±1	1.3±0.3
	7.3	220±10	17±1	0.6±0.2
	10.1	270±20	18±2	0

where $z=\gamma m_c T/B$ (m_c is in units of m_0), $\gamma=14.68$ T/K and $F_r=[2\cosh(2rz\delta T_D/T)+2\cos(2\pi r v)]^{1/2}$. Here $T_D=(T_D^{\uparrow}+T_D^{\downarrow})/2$ and $\delta T_D=(T_D^{\uparrow}-T_D^{\downarrow})/2$, where $T_D^{\uparrow\downarrow}$ are the Dingle temperatures for spin-up and spin-down electrons, respectively. Taking into account the relatively wide gap, $E_g\sim 0.3$ eV,[1] the effective g-factor for the Γ_6 conduction band can be expressed as $g_{\mathrm{eff}}=g^*+N_s\alpha\langle S_z\rangle/\mu_B B$, where $N_s=N_0\ y$ is the Mn concentration, α is the constant of the exchange interaction in the Γ_6 band, g^* is the exchange-dependent part of g_{eff} and $\langle S_z\rangle$ is the mean value of the projection of the Mn ion spin. The latter is connected with the magnetization, $M=N_s g\mu_B\langle S_z\rangle$, where $g\approx 2$ is the Landè factor. To take into account the generation of the magnetic clusters the magnetization is expressed usually with the phenomenological equation $M=gN_sS_0\mu_B B_s(\xi)$, where $\xi=gS\mu_B B/[k_B(T+T_0)]$, S_0 and T_0 are the effective spin and temperature parameters and $B_s(\xi)$ is the Brillouin function. The parameters S_0 and T_0 are determined by fitting the magnetization data with this expression (see Fig. 3). The values of $gS_0<5$ suggest formation of antiferromagnetic clusters, as in many other DMS.[6–8] Using values of S_0 and T_0, as well as those of $m_c(B)$ and T_D,[3] the SdH amplitudes were calculated with Eq. (1). From best fits of the calculated curves with the experimental data of the amplitudes $A_r(B)$ (see Fig. 2) the values of the parameters g^*, αN_0 and δT_D were determined. They are collected in Table I. Similar values of the exchange constant αN_0 have been found earlier in CZMA with $x=0.13$ and $y\leqslant 0.03$ (down to -0.3 eV)[9] and in some other DMS (0.28 eV in $Hg_{1-x}Mn_xSe$ and 0.27 eV in $Hg_{1-x-y}Cd_yMn_xTe$).[6,10] The values of g^* displayed in Table I agree with those in the parent alloys $(Cd_{1-x}Zn_x)_3As_2$ with comparable n_R and E_g($|g^*|=13$–37)[11] and in CZMA with $x=0.28$ and $y=0.02$ ($|g^*|=24$).[1] The SdH amplitudes (A_2) calculated at different values of δT_D are presented also in Fig. 2 along with the experimental data. Generally, A_2 is found to be more sensitive to the variation of δT_D than A_1. The dependence of both amplitudes on this parameter decreases when T is increased. It may also be concluded that δT_D decays rapidly with growing temperature (Table I). Such behaviour of δT_D agrees completely with that observed in other DMS.[5,9]

V. CONCLUSIONS

The analysis of the SdH effect in annealed CZMA crystals has been performed taking into account both the anomalous magnetic field dependence of the cyclotron mass and influence of the s-d interaction to the effective g-factor of the band electrons. The functions $m_c(B)$ obtained earlier on the annealed crystals[3] were used in the present study. The dependence of g_{eff} on temperature and magnetic field was introduced in the analysis of the SdH effect by using the results of the magnetization measurements made on the same CZMA specimens. By fitting the dependence of the first and the second harmonic amplitudes on the magnetic field values of the exchange-independent part of g_{eff}, g^*, the exchange constant in the conduction band, α, and the spin-dependent part of the Dingle temperature, δT_D, were determined (Table I). The values of g^*, α and δT_D are reasonable and agree with those determined in other DMS.[5–10] This gives evidence for coexistence of the two effects, the influence of the s-d interaction to the effective g-factor and the anomalous magnetic field dependence of the cyclotron mass, in the same material.

[1] R. Laiho, K. G. Lisunov, V. N. Stamov, and V. S. Zakhvalinskii, J. Phys. Chem. Solids **57**, 1 (1996).
[2] R. Laiho, A. V. Lashkul, E. Läderanta, K. G. Lisunov, V. N. Stamov, and V. S. Zakvalinskii, J. Phys. C **7**, 7629 (1995).
[3] R. Laiho, K. G. Lisunov, V. N. Stamov, and V. S. Zakhvalinskii, Cryst. Res. Technol. **31**, 605 (1996).
[4] *Annual Book of ASTM Standards*, (ASTM, Philadelphia, 1977), pt. 43, F. 76-73, p. 349–365.
[5] M. Vaziri and R. Reifenberger, Phys. Rev. B **32**, 3921 (1985).
[6] Z. Celinski, T. Mydlarz, and W. Zdanowicz, Acta Phys. Pol. A **71**, 239 (1987).
[7] W. Lubczynski, J. Cisowski, H. Bednarski, J. Voiron, J. C. Portal, and J. C. Picoche, Acta Phys. Pol. A **77**, 175 (1990).
[8] W. Dobrowolski, M. Ortenberg, A. Sandanen, R. P. Galazka, and R. Pantenen, Proc. of the 4th Intern. Conf. on Narrow Gap Semiconductors, Linz, Austria, 1981, p. 302.
[9] W. Lubczynski, J. Cisowski, J. Kossut, and J. C. Portal, Semicond. Sci. Technol. **6**, 619 (1991).
[10] W. Herbst, H. Pascher, F. Geist, M. Baran, T. Dietl, W. Dobrowolski, and R. Szymczak, Proc. of the 7th Intern. Conf. on Narrow Gap Semiconductors, Santa Fe, New Mexico, 1995, p. 80.
[11] E. K. Arushanov, A. A. Gubanova, A. F. Knyazev, A. V. Lashkul, K. G. Lisunov, and V. V. Sologub, Fiz. Tekh. Poluprovodn. **22**, 338 (1988) [Sov. Phys. Semicond. **22**, 208 (1988)].