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Low-temperature conductivity of $(Zn_{1-x}Mn_x)_3As_2$ solid solutions

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Abstract. The electrical conductivity of $(Zn_{1-x}Mn_x)_3As_2$ solid solutions with $0 \leqslant x \leqslant 0.13$ has been measured in the temperature interval of $4.3 < T < 200$ K. It is shown that the conductivity is of activational nature. The data are analysed with a model based on the disorder of Mn atoms over the vacancy-type sites of the metallic sublattice. At low temperatures the conductivity is determined by hopping of holes over the states in the lower Hubbard band whereas at high temperatures the holes are activated to states higher than the mobility edge of the upper Hubbard band. The observed complex dependence of the conductivity and the high-temperature activation energy on x are explained in terms of an increasing contribution of multiple spin-flip scattering of holes during their transitions between acceptor centres when x is increased.

1. Introduction

The alloys $(Zn_{1-x}Mn_x)_3As_2$, briefly ZMA, belong to a new class of semi-magnetic semiconductors (SMSCs) based on the II–V group compound Zn_3As_2. Three modifications of zinc arsenide are known (Arushanov 1980). In the high-temperature β-modification ($T > 945$ K) the unit cell has the FCC structure. In the low-temperature α-phase ($T < 945$ K) it consists of 16 fluorite cubes, each containing six metallic atoms and two vacancies. The vacancies are located at corner sites on one of the body diagonals of the cube. There are also four As atoms in the corners and facets of the fluorite cube. Between these phases there is the α'-modification, differing slightly from the α-phase. In the composition range $x \leqslant 0.15$ ZMA forms a substitutional solution of Zn_3As_2 and Mn_3As_2.

Zn_3As_2 and ZMA are p-type semiconductors with the energy gap $E_g \simeq 1$ eV (Arushanov 1980). The magnetic properties of ZMA are quite interesting, including a low-temperature spin-glass phase below $\simeq 4$ K (Denissen *et al* 1987) and a specific spin-freezing effect observable in low magnetic fields at temperatures around 200 K (Lähderanta *et al* 1992, Lashkul *et al* 1992, Chudinov *et al* 1992). The information about transport properties of ZMA is relatively meagre. The electronic conductivity and the galvano-magnetic properties of the alloy have been investigated by Kulbachinskii *et al* (1991). They assume that the carriers in ZMA are highly degenerate and that there is Kondo scattering of delocalized holes by magnetic moments associated with Mn. These assumptions may be applied to narrow-gap and zero-gap SMSCs, but they are doubtful in the case of ZMA for which $E_g \simeq 1$ eV and the low-temperature conductivity is of activational type as in Zn_3As_2 (Szatkovski and Sieranski 1990). Another possibility is to attribute the kinetic effects in ZMA and Zn_3As_2 to impurity centres and to properties of the impurity band.

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We have investigated the conductivity of ZMA for compositions $0 \leqslant x \leqslant 0.13$ in the temperature range of $4.2 < T < 200$ K. The data are analysed in the framework of the 'impurity mechanism'. The possible existence and the properties of an impurity band are also discussed.

2. Experimental details

Single crystals of ZMA were grown from stoichiometric amounts of Zn_3As_2 and Mn_3As_2 by using a modified Bridgman method. The Mn concentration of the crystals was in the range $0 \leqslant x \leqslant 0.13$. The compositions and homogeneity of the samples were analysed by x-ray and microprobe methods. The same material was used as in the previous study of the magnetic properties of ZMA (Lähderanta *et al* 1992, Lashkul *et al* 1992). The electrical conductivity measurements were made with the four-probe DC technique. The specimens were cooled in an He-exchange gas dewar and their temperatures were controlled with an accuracy of 0.02 K.

3. Experimental results

Our data indicate a strong dependence of the electrical conductivity, $\sigma(T)$, on the concentration of Mn in ZMA. As shown in figure 1 the samples with $x \leqslant 0.02$ have much higher conductivity than those where the Mn concentration is larger. Another important feature of the data is that when plotted in a logarithmic scale versus $1/T$, the temperature dependence of σ for compositions with $x < 0.05$ is almost linear, and for those with $x \geqslant 0.5$, exactly linear, in the temperature range $4 < T < 25$ K (called here the L or low-temperature region). Between 25 K and 75–100 K, depending on x, there is another region of conductivity (called the H or high-temperature region). According to the neutron diffraction data of De Vries *et al* (1989) ZMA crystals retain their α-Zn_3As_2 structure with x at least up to 0.135, Mn entering Zn sites substitutionally. Therefore the clear decrease of σ and the change in its temperature dependence above 25 K for $x \geqslant 0.05$ cannot be explained simply by a hypothetical change in the crystal structure when x is increased.

The results in figure 1 suggest that in all samples investigated $\sigma(T)$ has activational character below $T \simeq 100$ K. Assuming that in the H region the conductivity is determined by activation of holes to the mobility edge of the upper Hubbard band (UHB) we can write (Mott and Davies 1979, Shklovskii and Efros 1979)

$$\sigma_H(x, T) = \sigma_{0H}(x) \exp(-E_H/kT) \tag{1}$$

with

$$\sigma_{0H}(x) = \sigma_0 \exp(-\alpha_H/N_A^{1/3}(x)a(x)). \tag{2}$$

Here σ_0 is expected to depend weakly on x, α_H is a constant, $a(x)$ is the localization radius ($\simeq \hbar/(2m_{eff}E_H)^{1/2}$) in the UHB and N_A is the acceptor concentration. In the L region we can use expressions (1) and (2) by replacing E_H with E_L, α_H with α_L and defining $a(x)$ for the LHB. The values of the activation energies E_L and E_H for the L and H regions, respectively, are given in table 1. As can be seen from figure 2 E_L grows continuously while the Mn concentration is increased until a maximum (or saturation) is reached at $x = 0.10$. However, there is a singular point at $x = 0.01$ deviating downwards from this behaviour. At the same concentration E_H also has a low value.

The dependence of σ on x, measured at $T = 50$ K (curve 1) and at 6.7 K (curve 2) representing the H and L regions, respectively, is shown in figure 3. The two sets of data

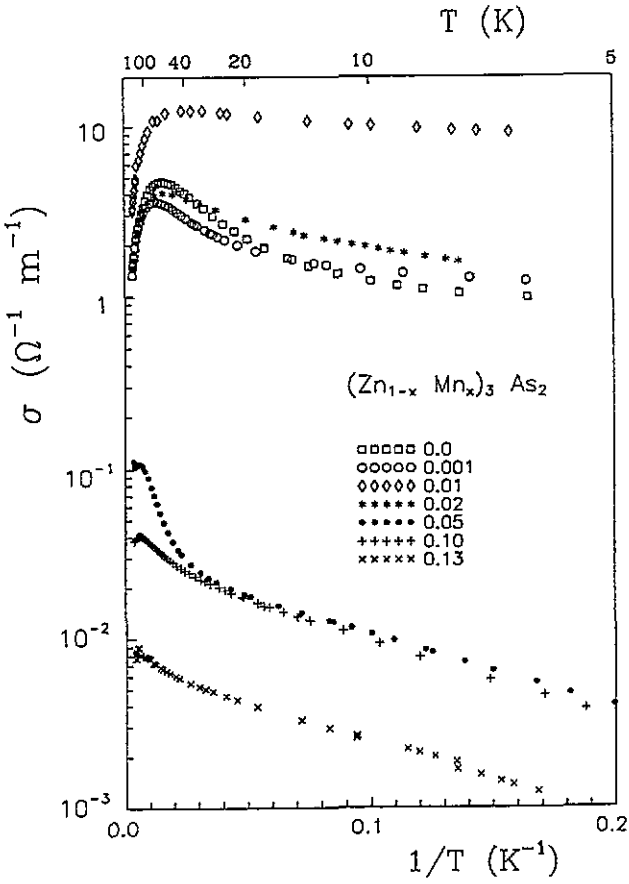

Figure 1. Temperature dependence of the conductivity of $(Zn_{1-x}Mn_x)_3As_2$ for different values of x.

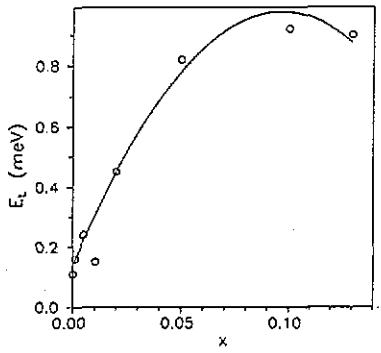

Figure 2. Dependence of the activation energy E_L on x. The full curve is a guide to the eye.

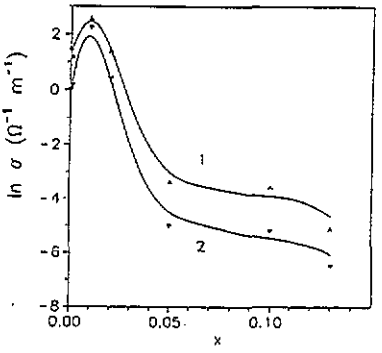

Figure 3. Compositional dependence of the conductivity measured at (1) $T = 50$ K and at (2) $T = 6.7$ K. The full curves are guides to the eye.

Table 1. Dependence of the activation energies E_H and E_L (in meV) on x.

x	0	0.001	0.005	0.01	0.02	0.05	0.10	0.13
E_H	2.3	1.8	0.9	0.3	1.1	7.5	2.4	1.7
E_L	0.11	0.16	0.24	0.15	0.45	0.82	0.92	0.90

are very similar having maxima around $x = 0.01$ and a region above $x = 0.05$ where σ decreases relatively slowly with increasing Mn concentration.

It is worth mentioning that Hall measurements have also been made in ZMA at a few selected temperatures (Zachvalinski 1992). The Hall concentration and mobility of holes at 77 K for $x = 0.01$ and 0.02 are $p_H = 5 \times 10^{17}$ cm^{-3}, $\mu_H = 2$ cm^2 V^{-1} s^{-1} and $p_H = 2 \times 10^{17}$ cm^{-3}, $\mu_H = 0.1$ cm^2 V^{-1} s^{-1}, respectively.

The data of Kulbachinskii *et al* (1991) indicate an approximately logarithmic dependence of the resistivity ρ on T over a limited range of T depending on the concentration of Mn in the sample. This observation was used to attribute the increase of the resistivity when cooling the sample to the Kondo effect (Kondo 1964). In the H region our data can be represented as $\ln(\sigma) \propto 1/T$ although in some interval ($\simeq 30$ K wide) on the low-temperature side of the maxima of σ in figure 1 the formula $\rho \propto -\ln(T)$ might be approximately used. Below 20 K (L region) the conductivity of all our samples can be represented much better with $\ln(\sigma) \propto 1/T$. This is particularly clear in the specimens with $0.05 \leqslant x \leqslant 0.13$. To account for these features we have developed a model of conductivity without using the Kondo effect. This will be discussed in the next section.

4. Discussion of results

Neutron diffractograms of ZMA have revealed an additional weak diffusive maximum (De Vries *et al* 1989). It was suggested that this peak may result from non-random distribution of Mn atoms (De Vries *et al* 1989). On the other hand, its diffraction angle is compatible with a disorder of the systematic stacking of the two vacancies in the fluorite cubes shown in figure 4(*a*) (De Vries 1989). This case would lead to generation of impurity centres and is therefore an attractive choice for the discussion of our conductivity data. Since the origin of the diffusive scattering is not completely clear and the explanations given for it are interrelated we have to emphasize the somewhat speculative nature of the approach presented below.

The disorder shown in figure 4(*b*) is connected with generation of a pair consisting of a bivalent donor (M_1 in $V_{CG}^{(1)}$) and a bivalent acceptor ($V_M^{(1)}$), where M, V_{CG} and V_M represent the metal atom, the vacancy in an ideal structure and the vacancy in the case of disorder, respectively. M_1 in $V_{CG}^{(1)}$ cannot restore its broken bonds with M_5 and M_6 (because the angles of the bonds (M_1–M_5, M_2–M_5) and (M_1–M_6, M_4–M_6) would be two times smaller), whereas M_5 and M_6 can create a new bond with each other at the same bond angle but with another bond distance. However, change in the distance leads to an additional distortion of the metallic cube and creation of a potential pit deep enough to contain two superfluous valence electrons of M_1. So, even with a small disorder of this type we obtain a great amount of highly compensated acceptors with two levels (otherwise such disorder would not lead to a p-type semiconductor), the compensation being incomplete due to background impurities. We denote the atoms occupying the sites of the ideal sublattice the 'lattice atoms' and those causing the disorder of vacancies the 'vacancy-type atoms'.

The acceptor concentration N_A does not coincide with that of the vacancy-type atoms, because there must be other types of disorder, differing from that shown in figure 4(*b*). For example, in the case of disorder shown in figure 4(*c*), the vacancy-type atom is able to restore its bonds and N_A may differ from the total concentration of the vacancy-type atoms by a factor of $b < 1$. The process of disordering is expected to have a sharp maximum near the $\beta \rightarrow \alpha'$ phase transiton ($T = 945$ K) resulting in a serious reconstruction of the unit cell in zinc arsenide and in ZMA.

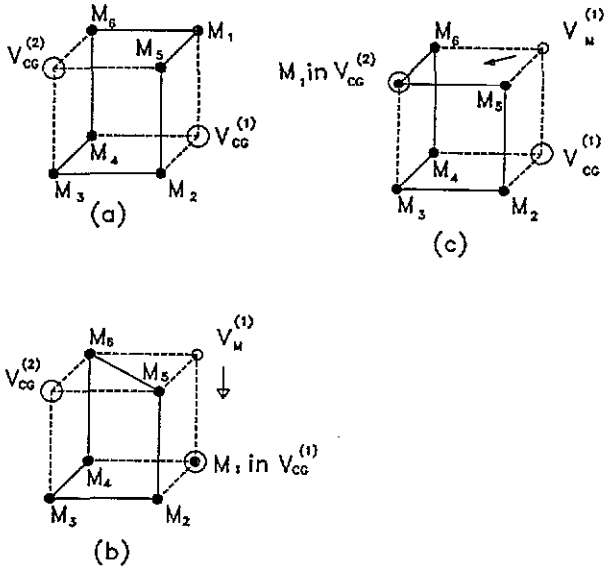

Figure 4. Model of vacancy-type disorder in $(Zn_{1-x}Mn_x)_3As_2$. (*a*) Ideal ordering of vacancies. (*b*) Disorder leading to appearance of a metallic atom M_1 with broken bonds. (*c*) Case of disorder when M_1 restores its bonds with metallic atoms.

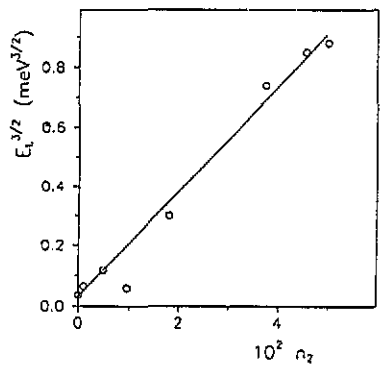

Figure 5. Dependence of $E_L^{3/2}(x)$ on $n_2(x)$. The full line is a fit to (6).

Two impurity bands of the Hubbard type are likely to result from broadening of the two acceptor levels. Hence the activational conductivity in the L region is probably due to hopping conductivity over the strongly localized states of the LHB, the Fermi level being deep in it due to the high compensation. The transition to hopping conductivity on lowering the temperature has been observed also in undoped Zn_3As_2 (Szatkovski and Sieranski 1990). In the H region the activational conductivity must be attributed to activation of holes to delocalized states higher than the mobility edge E_{me} of the UHB. It is confirmed by the very low values of the Hall mobility completely excluding the activation of the holes to the states of the valence band (VB).

The relative concentrations of the vacancy-type Zn and Mn atoms, $n_1 = N_1/N$ and $n_2 = N_2/N$ respectively, can be obtained by minimizing the free energy $F = E - TS$ (N is the concentration of Zn atoms in Zn_3As_2 and $N/3$ is that of the vacancies), considering that the exchange interaction between the Mn atoms influences mostly the ordering but not the concentration of the vacancy-type Mn atoms. The configurational entropy is $S = k\ln(W_1W_2)$, where

$$W_i = \{(N\xi_i)!/[N_i!(N\xi_i - N_i)!]\}(N/3 - N_j)!/[N_i!(N/3 - N_i - N_j)!] \qquad i \neq j = 1, 2. \tag{3}$$

The configurational energy can be expressed as $E = E_1N_1 + E_2N_2$, where E_1 and E_2 are the mean activation energies of the vacancy-type Zn and Mn atoms, respectively. Thus we obtain the following set of equations

$$n_i^2(1 - 3n_i)/[(\xi_i - n_i)(1 - 3n_i - 3n_j)^2] = \tfrac{1}{3}A_i \qquad i \neq j = 1, 2 \tag{4}$$

where $\xi_1 = 1 - x$, $\xi_2 = x$, and $A_i = \exp(-E_i/kT)$ with $i = 1, 2$. In (4) we are interested in the dependences of N_1 and N_2 on x. Most of the defects are generated at the phase transition temperature $T(\beta \rightarrow \alpha')$. We can assume that the concentrations of N_1 and N_2 are frozen in at that temperature and depend only on x. The constants A_1 and A_2 are taken

at low temperatures equal to their values at $T(\beta \to \alpha')$. x is relatively small; therefore we can solve (4) to the second order in x,

$$n_i(x) = \sum_{k=0}^{2} n_{ik} x^k \qquad i = 1, 2 \qquad n_{20} \equiv 0. \tag{5}$$

From the effect of sublimation of Zn_3As_2, described e.g. by Lazarev *et al* (1978), we estimate that $E_1 \simeq 1$ eV, which is a typical value of the activation energy of defects in metals. Assuming that n_2 at $x_0 = 0.135$ is equal to the intensity of the diffusive neutron scattering peak in ZMA divided by that of the Zn maximum (this fraction at $x = x_0$ is equal to $\simeq 0.04$ (De Vries *et al* 1989)) and substituting (1) into (4), we obtain $n_{12} \simeq 1$. Using the values of $E_1 \simeq 1$ eV and $n_2(0.135) = 0.04$, we get $A_1 \simeq 5 \times 10^{-6}$, $n_{10} \simeq 10^{-3}$, $n_{11} \simeq 4 \times 10^{-3}$, $n_{22} \simeq -5$, $A_2 \simeq 0.6$ and $n_{22} \simeq 2 \times 10^{-3}$. The value of A_2 corresponds to $E_2 \simeq 0.04$ eV which is much lower than E_1 and is comparable with the energy of the exchange interaction of Mn atoms with their nearest neighbours (Denissen *et al* 1987).

The values of n_{1i} $(i = 1, 2)$ are much smaller than those of n_{2i}. Hence the acceptor concentration $N_A(x)$ is determined for $x > 0$ only by the vacancy-type Mn atoms. It can be seen that the values of n_{21} and n_{12} are found with an accuracy sufficient to obtain the relationship $n_2(x) \simeq x - 5x^2$ which determines the main character of the variation of $N_A(x)$ with growing x.

A possible explanation for the growth of $E_L(x)$ with increasing x (figure 2) is the polarization of the magnetic moments by localized charge carriers (Mott and Davies 1979). This effect must lead to a rapid increase of resistivity when the temperature is lowered. For instance in EuO doped with excess Eu the decrease of $\sigma(T)$ is about three orders of magnitude in the temperature range of 2–10 K (Von Molnar 1970). Such a polarization also leads to a large negative magnetoresistance (MR) as observed in EuS:La (Methfessel and Mattis 1968) where the decrease of the resistivity is about three orders of magnitude in the interval of 0–14 kOe at 20 K. These effects are not observed in ZMA (Zachvalinski 1992). Although in ZMA a large negative MR at low temperatures may be cancelled by a positive MR typical of materials exhibiting hopping conductivity (Methfessel and Mattis 1968), the following estimation shows that the increase of E_L with x can hardly be connected with the polarization of the magnetic moments by localized carriers. A localized charge carrier is able to polarize only an amount of spins $n_p \sim \delta E_L / J_1$, where δE_L is the polarizational part of E_L and J_1 is a value of the exchange interaction between the carriers and the magnetic moments of Mn. It is required for the polarization of the spins that J_1 is comparable with the exchange interaction between the Mn atoms ~ 10 meV (Denissen *et al* 1987). For the maximum value of $\delta E_L \sim 1$ meV we get a small number of $n_p \sim 0.1$.

Another possibility to explain the dependence of E_L on x may be connected with large-scale fluctuations (LSFs) of x. The growth of the activation energy of hopping conductivity due to this reason has been observed in $Ge_{1-x}Si_x$ (Shlimak *et al* 1977). However, the influence of LSFs on the disorder of the acceptor levels in ZMA is suppressed by the correlated distribution of the vacancy-type Mn atoms, consisting of no less than half of the Mn atoms in an alloy with x up to $\simeq 0.1$.

It was supposed above that the acceptor states of the LHB are strongly localized and highly compensated. In this case $E_L(x) \sim N_A^{1/3}(x)[1 - K(x)]^{-1/3}$ with $N_A(x) = bNn_2(x)$ (Shklovskii and Efros 1979). Writing

$$1 - K(x) = [1 - K(0)]N_A(0)/N_A(x)$$

we obtain an expression

$$E_L(x) = E_L(0)(1 - (Nb/N_A(0))n_2(x))^{2/3}. \tag{6}$$

This formula predicts a linear dependence of $E_L^{3/2}(x)$ on $n_2(x)$, in good agreement with the experimental results in figure 5, excluding the point at $x = 0.01$. This value of x corresponds to the minimum of $E_L(x)$ and $E_H(x)$ and the maximum of $\sigma(x)$ both in the L and H regions allowing us to suggest that at $x \simeq 0.01$ the system is close to the metal–insulator transition (MIT) i.e. E_{me} comes close to E_F. Close to the MIT, equation (6) is not valid (Mott and Davies 1979), leading to a non-linear dependence of $E_L^{3/2}(x)$ on $n_2(x)$. The similar behaviour of $\sigma(x)$ in the low- and high-temperature regions supports the idea that in both cases the conductivity is connected with transitions of holes between acceptors. In such a situation $\sigma(x)$ is determined mostly by the degree of overlap between the acceptor states (Mott and Davies 1979, Shklovskii and Efros 1979). Since $N_A(x)$ grows with increasing x there must be a corresponding growth of the conductivity. This is observed up to $x \simeq 0.01$. At $x > 0.01$ $\sigma(x)$ shows an anomalous behaviour, decreasing with growing x. We assume that this decrease is connected with multiple spin-flip scattering of holes during their transitions between acceptors. The following mechanism can be proposed. Due to the presence of a local magnetic field H_i the spin-flip scattering has inelastic character, the energy loss of a hole during one transition being $\delta E \sim n_s \delta E_1$ where n_s is the number of scatterings per transition and $\delta E_1 \sim \mu_B H_i$ is the change of energy per scattering event. If we have $\delta E \sim E_0$ (mobility edge at $x = 0$) this scattering leads to an increasing binding energy of the hole at the acceptor. This in turn leads to the decrease of the localization radius, a, and therefore to a decreasing overlap of the acceptor states resulting in smaller values of $\sigma(x)$. In the case of localized states the scattering amplitude can be estimated by using the expression for the scattered wave (Spivak *et al* 1985) and a first-order correction to the wavefunction. This gives $\mu_s \sim aJ_1/E_0$. For the H region we estimate that $\mu_s \sim R_A J_1/E_0$, $E_0 \sim E_H(0) \sim 1$ meV and $R_A \sim (bNn_2)^{-1/3}$ (mean distance between acceptors). Then, for $n_s \sim \mu_s^2 R_A N_s$ where $N_s \sim Nx \sim Nn_2$ is the concentration of the scattering centres, we obtain $\delta E \sim (J_1/E_0)^2 \mu_B H_i/b$. For $x \sim 0.1$, $H_i \sim 10^2$ Oe (Lashkul *et al* 1992) and $E_0 \sim E_H(0)$. Then $\delta E \sim E_0$ leads to $(J_1/E_0)^2/b \sim 10^3$ which can be easily fulfilled with realistic values of $J_1 \sim 1$–10 meV and $b \sim 10^{-1}$–10^{-3}. From figure 1(*b*) it follows that the variation of $a(x)$ is rapid for $x < 0.05$ and slower in the region of $x > 0.05$. Because $n_2(x)$ behaves in the same way the Mn vacancy atoms probably play the leading role in the spin-flip scattering.

Two opposite tendencies are connected with the growth of the Mn concentration: (i) increase of the overlap of the acceptor states due to growth of $N_A(x)$ and (ii) decrease of it due to the decrease of $a(x)$ by the spin-flip scattering. It leads to a maximum in $\sigma(x)$ and a minimum in $E_H(x) = E_{me} - E_F$, both of them taking place near $x = 0.01$ where the tendencies (i) and (ii) are probably comparable. A further increase of x leads to an enhancement of the spin-flip scattering rate resulting in a rapid decrease of $\sigma(x)$ and increase of $E_H(x)$ in the range of $0.01 < x < 0.05$. The slow decrease of $a(x)$ for $x > 0.05$ also causes the slow decrease of $\sigma(x)$, whereas $E_H(x)$ falls rapidly for $0.05 < x < 0.1$. As has been mentioned above, the main role in the disordering of the acceptor levels is connected with the disorder of the next-nearest neighbours in the subsystem of the vacancy-type Mn atoms. The above considerations of $E_L(x)$ confirm this conjecture. Therefore the decrease of $E_H(x)$ for $x > 0.05$ must be attributed to a transition of the vacancy-type Mn atoms into an ordered state, E_{me} being very sensitive to the degree of disorder (Mott and Davies 1979). So we see that the behaviours of $\sigma(x)$ and $E_H(x)$ are determined by changes of the magnetic subsystem near $x = 0.05$. Freezing of the magnetic moments at $T_f \simeq 200$ K (H region) has been reported by Lashkul *et al* (1992).

5. Conclusions

We have measured the electronic conductivity of $(Zn_{1-x}Mn_x)_3As_2$ solid solutions with $0 \leqslant x \leqslant 0.13$. The results have been discussed in terms of a model based on disorder over vacancy-type sites of the metallic sublattice. The behaviour of the low-temperature activation energy following from this model is in good agreement with the experiment, as well as with the complex dependence of the high-temperature activation energy and anomalous dependence of the conductivity on x.

The growth of II_3V_2-type compounds from the vapour phase takes place below the $\alpha' \rightarrow \beta$ phase transition resulting in a much smaller amount of defects (Arushanov 1980). Therefore the comparison of the present results with those obtained with crystals grown from the vapour phase may provide valuable information about the validity of the impurity model applied. Another interesting question is the behaviour of the electrical conductivity of $(Cd_{1-x}Mn_x)_3As_2$ (CMA). This alloy is isomorphic to ZMA (De Vries *et al* 1989) and also exhibits a magnetic freezing phenomenon at high temperatures (Laiho *et al* 1992). Therefore there is a possibility of generation of similar types of disorder in these two materials. In CMA we may expect metallic conductivity by lowering the temperature. This allows the influence of the spin-flip scattering on the conductivity to be studied in a more straightforward way.

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