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Spin-glass like behaviour of $(Zn_{1-x}Mn_x)_3As_2$

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Magnetic properties of the diluted magnetic semiconductor $(Zn_{1-x}Mn_x)_3As_2$ (ZMA) have been investigated for compositions $0 \le x \le 0.13$ and temperatures $4\ K \le T \le 300$ K. Several phenomena typical to spin-glasses were observed: history dependence, maxima in the functions $\chi_{dc}(T)$ and $\chi_{ac}(T)$ and smearing out of the maximum of $\chi_{dc}(T)$ in strong fields. For samples with $x = 0.10$ and 0.13 our data give magnetic freezing temperatures around 200 K which is unusually high for diluted magnetic semiconductors.

$(Zn_{1-x}Mn_x)_3As_2$, shortly ZMA, is one of the diluted magnetic semiconductors (DMS). Usually for DMS a spin-glass transition is observed at low temperatures [1]. Also ZMA undergoes a spin-glass transition and for the concentrations $0.055 \le x \le 0.14$ the freezing temperature is reported to be in the range $0.035 \le T_{SG} \le 4.3$ K [2].

We have investigated ac- and dc-magnetic properties of ZMA by using weak magnetic fields and a wide temperature range. The results obtained for compositions $0.10 \le x \le 0.13$ indicate spin-glass like freezing of the magnetic moments around 200 K.

Single crystals of ZMA used in this work were grown by the modified Bridgman method: Slow cooling of a sample over which a temperature gradient is established. Increasing the relative manganese concentration to more than 0.15 leads to growing inhomogeneities with the second phase $ZnMn_2As_2$ as shown by metallographic, thermographic and X-ray analyses [3]. Monocrystallicity, composition and homogeneity of crystals obtained were controlled by X-ray techniques. All crystals had the α-phase Cd_3As_2 structure with space group $I4_1cd$ at 300 K.

The ac-magnetic measurements in the temperature range 77–300 K were performed with a mutual inductance bridge using the driving frequency of 320 Hz and deriving fields of 0.7 Oe for samples with $x = 0.10$ and 0.13, and 3.5 Oe for those with $x = 0.05$.

The dc-magnetic measurements were performed with a SQUID magnetometer. Due to the high sensitivity of the magnetometer we were able to detect small signals using weak magnetic fields which is essential when investigating spin-glasses (see below). The temperature was measured with the accuracy of 0.2% by using a carbon-glass thermometer. The external magnetization field was produced by a superconducting magnet.

Pure Zn_3As_2 showed diamagnetic behaviour with $\chi = -2.2 \times 10^{-7}$ emu/gOe. This susceptibility value agrees with an earlier result [2]. ZMA containing a small amount of Mn has paramagnetic properties above 4 K as can be seen from the dc-susceptibility data for concentrations between $0.001 \le x \le 0.02$ (fig. 1). In fig. 1 are shown susceptibilities of both the zero field cooled (ZFC; open symbols) and field cooled (FC; closed symbols) samples. The history dependence of the dc-magnetization of the sample with $x = 0.02$ can be observed as a small difference between the ZFC and FC curves below 20 K. For the sample with $x = 0.05$

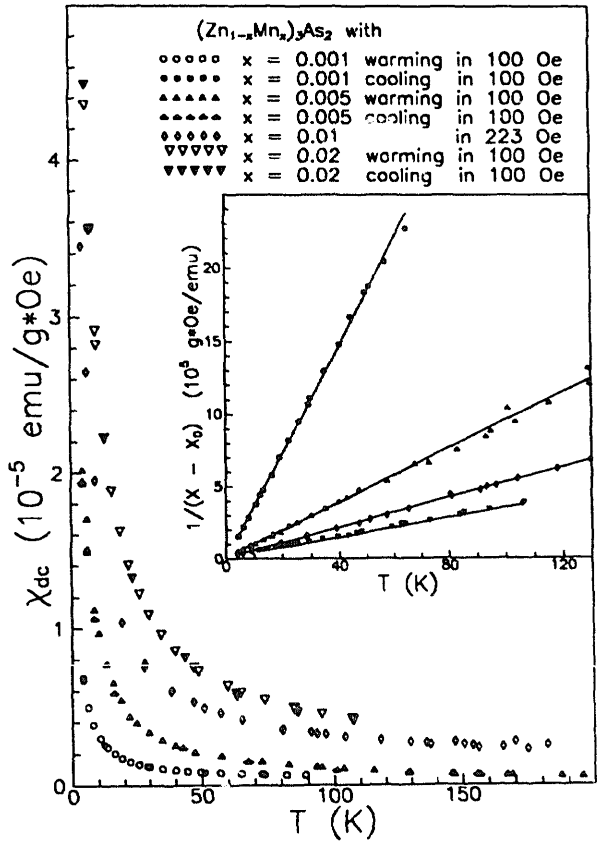

Fig. 1. DC-susceptibility of ZMA for $x \le 0.05$. Open and closed symbols indicate the ZFC- and FC-samples, respectively. The inset shows a Curie–Weiss fit of the results.

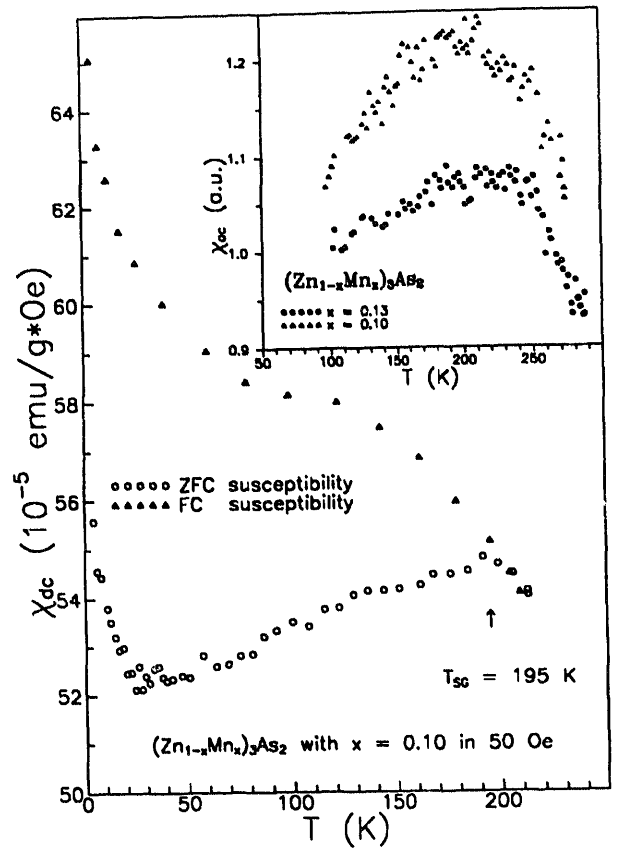

Fig. 2. Spin-glass like behaviour of ZMA $x = 0.10$. The maximum in the ZFC curve at $T_f = 195$ K can easily be seen. The inset shows the ac-susceptibilities of the $x = 0.10$ and 0.13 samples, measured at the frequency of 320 Hz and the driving field of 0.7 Oe.

this difference is more distinct, but by increasing the measuring field it disappears.

In fig. 2 is shown the dc-susceptibility of the $x = 0.10$ sample measured at 50 Oe. The history dependence is extremely large and a maximum can be seen in the ZFC curve.

In the inset is presented the temperature dependence of the ac-susceptibility (χ_{ac}) for the samples with $x = 0.10$ and 0.13. We could use only a small amount of the sample material and therefore the scattering of the experimental points is large. Anyhow a maximum can be seen but it is not very sharp. The sample with $x = 0.05$ indicated the Curie–Weiss behaviour down to 80 K which was the temperature limit of this experiment.

Spin-glass like properties can be seen as the history dependence of the magnetization already in the samples with $x = 0.02$ and 0.05, although their behaviour is approximately paramagnetic. In addition to this a good indication of the spin-glass phase is the appearance of the maximum in the ZFC susceptibility curve as shown in fig. 2. All these properties disappear in magnetic fields of a few kilogauss and this is an obvious reason why they were not seen earlier [2]. We attribute also the maximum in the $\chi_{ac}(T)$ curves to the freezing of moments. Therefore, according to our results, the samples with $x = 0.10$ and 0.13 show spin-glass behaviour and the sample with $x = 0.05$ is near the border line of the paramagnetic to the spin-glass transition. From the positions of the maxima in $\chi_{ac}(T)$ we estimate that the freezing temperature T_f for the samples with the Mn concentrations $x = 0.10$ and 0.13 are (205 ± 15) and (230 ± 15) K, respectively. The dc-measurement at 50 Oe gave $T_f = 195$ K for the sample with $x = 0.10$.

The existence of two spin-glass phase in the same material in two totally different temperature-regions is quite unusual. A possible explanation would be the existence of two or more types of magnetic clusters. An alternative mechanism might be frustration or some other change in the antiferromagnetic ordering as found by neutron scattering in $Cd_{1-x}Mn_xTe$ [4].

We have investigated magnetic properties of $(Zn_{1-x}Mn_x)_3As_2$ in a wide range of temperatures and applied fields. Unusual complexity of the spin-glass behaviour of this material has been observed: In addition to the already known low temperature spin-glass phase [2] a new freezing phenomenon of spins is found at about 200 K. The anomalies of the ac- and dc-susceptibilities corresponding to this phenomenon smear gradually out when the measuring field is increased beyond a few kilogauss. The obtained data are in favour of freezing out of spin clusters but before drawing final conclusions additional information, as obtainable by polarized neutron experiments, would be desirable.

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