PHASE MAGNETIC TRANSFORMATIONS IN MnAs1-xSbx ALLOYS

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PHASE MAGNETIC TRANSFORMATIONS IN $MnAs_{1-x}Sb_x$ ALLOYS

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Abstract. – The structure and magnetic properties of single crystals of a continuous series of $MnAs_{1-x}Sb_x$ solid solutions have been studied in the temperature range from 150 to 600 K. It is shown that the mechanism and kinetics of the phase transformations in the above mentioned system is determined by the ordering of magnetoelastic static lattice distortions. From the measured data, the phase diagram of transformations in the $MnAs_{1-x}Sb_x$ single crystals has been plotted.

1. Introduction

Research into MnAs involves, first of all, the elucidation of mechanism and kinetics of the first order magnetic phase transition at $T_u = 313$ K, an investigation of the anomalous behaviour of magnetic susceptibility above T_u and finally the influence of various additions [1-3]. Of special interest, in this latter respect, is the study of the effect of Sb additions on the above mentioned phase transitions, which leads to the increase in the unit cell volume. Earlier phase transition studies on polycrystals of $MnAs_{1-x}Sb_x$ alloys revealed the decrease in phase transition temperature T_u with increasing Sb concentration [2, 3].

2. Experimental

Single crystals of the $MnAs_{1-x}Sb_x$ solutions were grown by the Stockbarger-Bridgman method in double unsealed quartz ampoules. The change of lattice translation periods was determined in X-ray analysis in the range from 150 to 600 K, the atomic coordinates were calculated and polar diagrams of squariance dynamic displacements of atoms u_s^2 were plotted. The method of X-ray structure analysis was described in [4]. The magnetic measurements were limited to the study of the magnetic anisotropies and temperature dependences of remanent magnetic induction for the single crystals.

The measurements of the temperature dependences of the lattice parameters of the single crystals has shown that a first order transition exists at T_u up to the composition $MnAs_{0.5}Sb_{0.5}$. This transition is accompanied by a discontinuity in the change of the crystal lattice volume, whose value reduces from $\Delta V / V_0 = 0.022$ for MnAs to $\Delta V / V_0 = 0.005$ for $MnAs_{0.5}Sb_{0.5}$.

The static lattice deformation has been followed through a determination of the temperature dependences of manganese atoms displacements from the equilibrium positions in the NiAs structure which are illustrated in figure 1. From these data, it follows that the crystal structure of MnAs in the ferromagnetic state at 293 K, for example, is characterized by the availability of uniaxial displacements for Mn atoms at $z_{Mn} = 0.015$ along [001] (shifts from [001] for the all

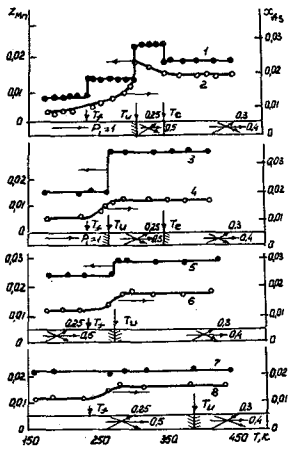
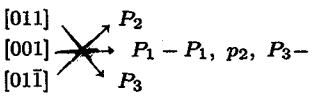


Fig. 1. – Atomic static displacement of Mn–z_{Mn} and As–x_{As} from equilibrium positions NiAs-type structure *vs.* temperature in the MnAs (1.2), $MnAs_{0.05}Sb_{0.95}$–$MnAs_{0.2}Sb_{0.8}$ (3.4), $MnAs_{0.3}Sb_{0.7}$–$MnAs_{0.5}Sb_{0.5}$(5.6), $MnAs_{0.2}Sb_{0.8}$(7.8).



probabilities of Mn atom displacements in the [001], [011], [01$\bar{1}$].

compositions is 8°) and arsenic $x_{As} = 0.014$ in [100] (MnP structure). As the temperature is lowered the atomic displacements decrease: those of arsenic reducing down to $x_{As} = 0.008$, and those of manganese showing a jump to $z_{Mn} = 0.01$ at $T_f = 240$ K. It is shown that the manganese atomic displacements determine the direction of easy magnetization in single crystal to lie in (100). At the first order phase transition, we observed both a growth in the values of atomic displacements and the disorientation of the manganese atomic displacements and the disorientation of the manganese atomic displacements from being uniaxial along [001] to include displacements along [011] and [01$\bar{1}$] with the ratio of probabilities of atoms displacements $P_1 : P_2 : P_3 = 0.5 : 0.25 : 0.25$. On raising the temperature further some decrease in the atoms displacements is observed. The arsenic atom displacements fall gradually to the value of $x_{As} = 0.019$,

whereas the manganese atom displacements exhibit a sudden reduction at $T_c = 350$ K to the value $z_{Mn} = 0.02$. Simultaneously, a further disorientation of the manganese atom displacements along [001], [011], [01$\bar{1}$] occurs to the probabilities $P_1 : P_2 : P_3 = 0.4 : 0.3 : 0.3$.

For single crystals with Sb concentration $0 \leq x \leq 0.2$ the temperature dependences of static atomic displacements (Fig. 1, curves 3, 4) are similar to those for equiatomic MnAs. A striking feature of these alloys is the increase in the atomic displacements due to the increasing quantity of structural defects. On increasing the Sb content in the range $0.3 \leq x \leq 0.5$ (Fig. 1, curver 5, 6), further atomic displacements were observed due to the increasing imperfection of the crystal lattice. The disorder of structure defects defines the disordering character of the manganese atomic displacements in the whole investigated temperature range. The phase transition at T_u in this case is accompanied by an insignificant change of values of manganese and arsenic atoms displacements as well as the degree of disordering. In single crystals with Sb content $x = 0.5$ the phase transition at T_u is followed by anomalous change of the thermal expansion coefficient and some disordering of manganese atoms displacements (Fig. 1, curves 7, 8) for $MnAs_{0.2}Sb_{0.8}$. For manganese antimonide the magnetoelastic atomic displacements are small in value $x_{As} = 0.006$, $z_{Mn} = 0.008$. In this case the ordering of displacements at $T_u = 480$ K does not lead to destruction of the ferromagnetic state.

Magnetic investigations of the $MnAs_{1-x}Sb_x$ system single crystals consisted in measuring the magnetic anisotropy and remanent magnetic induction B_r in (100), their variation with temperature in the range from 100 to 700 K.

3. Conclusions

A phase transformation diagram has been plotted for the $MnAs_{1-x}Sb_x$ system (Fig. 2) from the measured results for the disordering of atomic shifts T_u and those of temperature of the transition to the paramagnetic state T_c. As may be seen from the diagram, the ferromagnetic ordering of magnetic moments (F) in (100) is a characteristic of all the single crystals below the phase equilibrium line T_u. For single crystal containing $0.2 \leq x \leq 0.9$ of Sb, a uniaxial magnetoelastic crystal lattice deformation arises (MnP-type structure). In the middle range of compositions $0.2 < x \leq 0.8$, the structure defects specify the disordered character of manganese atoms displacements from their equilibrium positions in the NiAs structure.

The transition from the uniaxial atomic displacements to a disordered one with composition, when the static atomic displacements to the mean square displacements $\overline{u_s^2}$ are included, can be considered as a rise in lattice symmetry.

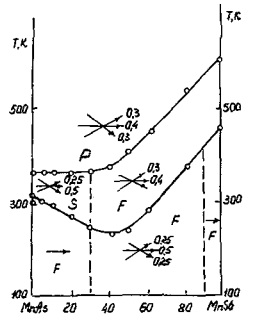


Fig. 2. – Phase diagram of transformations in the $MnAs_{1-x}Sb_x$ system single crystals. P - paramagnetic, F - ferromagnetic, S - layered magnetic.

The phase transition on the T_u line for single crystals with $x \leq 0.2$ is accompanied by a change in the amplitude of atomic displacements and their disordering from uniaxial to being along three directions [001], [011], [01$\bar{1}$]. As has been mentioned, in single crystals with Sb concentration $x \leq 0.2$ the direction of the magnetic moment in (100) is unambiguously determined by the displacements of Mn atoms in the lattice. A similar correlation allows one to suppose that the magnetic structure of single crystals with Sb concentration $x \leq 0.2$ in the temperature range between the lines T_u and T_c corresponds to ferromagnetic ordering of moments in (100), though in the neighbouring layers their direction may be distinguished by an arbitrary angular multiple of 60° (S). The destruction of the layered magnetic structure (S) at the transition to paramagnetic state (P) occurs in this case on the T_c line.

For single crystals with Sb content $x \geq 0.9$, the transition from the state with uniaxial displacements of Mn atoms to the state of disordered shifts on the T_u line does not lead to the destruction of the magnetic state due to small values of these shifts $x_{As} = 0.006$, $z_{Mn} = 0,008$. The transition to T_u is revealed by the change of magnetic anisotropy – transition to magnetic structure of easy plane type. In the range of composition $0.2 < x \leq 0.8$ the static displacements of atoms are disordered by the defects of crystal structure. In this connection the MnP-type structure with disordered displacements can be considered as practically stable in the whole investigated temperature range. As a result, the change of magnetization with temperature for middle compositions is described by the Brillouin curve.

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