Antiferromagnetic Ordering Arising in $\text{Mn}_{19.8-x}\text{Fe}_x\text{Sn}_{0.2}$ Alloys with the $\beta$-Mn-Type Structure


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Abstract—The magnetic state of the $\text{Mn}_{19.8-x}\text{Fe}_x\text{Sn}_{0.2}$ alloys is revealed and the mechanisms of changing their properties depending on the iron content are determined.

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Paramagnetism of $\beta$-manganese is a subject of numerous discussions, and the alloys based on $\beta$-Mn attract the attention of many researchers [1–6]. In particular, this interest is associated with the local environment of Mn atoms and the magnetic state of the $\beta$-Mn diluted alloys. The allotropic modification of $\beta$-Mn crystal has a cubic structure of type A13 and space group P4$_3$32. Twenty Mn atoms forming the unit cell are distributed in two crystallographically nonequivalent positions 8c and 12d [7, 8], with their atomic radius in position 12d being smaller than that for any electron configuration in the 8c position.

From the results of NMR studies of $\beta$-Mn alloys, it was concluded [9] that, in most positions, Mn atoms behave almost independently in magnetic respect and only the 12d position is magnetic (it was shown that only the atoms located in the 12d sites are responsible for the occurrence of the spin glass states in this system, whereas the atoms occupying the 8c sites remain nonmagnetic). Accordingly, in paramagnetic $\beta$-Mn, the atoms at sites 12d are the main contributors into antiferromagnetic spin fluctuations. The atoms in 12d positions form a grid consisting of regular triangles. Each atom in this position is connected with six other atoms in the 12d position, so that three triangles have a common apex. Such a geometry in conjunction with the antiferromagnetic Mn–Mn exchange interaction result in frustrated coupling, which prevents the formation of a certain equilibrium antiferromagnetic structure. As a consequence, no stable long-range magnetic order can be established in $\beta$-Mn [9].

A number of studies [4–6, 9–12] are devoted to the structure and properties of the $\beta$-Mn-type systems with small additions of nonmagnetic (Sn, Al) or magnetic (Fe) atoms. The data of X-ray diffraction analysis suggest that Al substituents almost exceptionally (90%) occupy the magnetic 12d sites [9]. A similar conclusion was drawn for the Sn atoms from the analysis of the Mossbauer spectra of $\beta$-$\text{Mn}_{19.3-x}\text{Sn}_{0.7}\text{Fe}_x$ alloys [6]. At the same time, it was shown that iron atoms have a strong preference for the nonmagnetic 8c positions. Such a preferred substitution leads to a different local environment of Mn atoms in $\beta$-$\text{Mn}_{1-x}\text{M}_x$ alloys depending on substituting element M and, as a result, to a strong dependence of the material properties on M.

Analyzing the Mossbauer spectra and neutron diffraction data, the authors of [3, 4, 13, 14] came to a conclusion that antiferromagnetic ordering appears in these alloys when Mn atoms are substituted by Sn atoms. The same results were obtained in [15] for $\text{Mn}_{16-x}\text{Fe}_x\text{Sn}_{16}$, $\text{Mn}_{19.9-x}\text{Fe}_0\text{Sn}_x$, and $\text{Mn}_{16-x}\text{Fe}_x\text{Sn}_{16}$ alloys containing both Sn and Fe atoms simultaneously. Nevertheless, some specific features of the magnetic behavior of Mn-based alloys remain unclear. In particular, the magnetic properties of some quasi-binary systems in relation to their composition are yet to be studied and their magnetic state is still unknown. Some literature data come into conflict. For example, the absence of antiferromagnetic order was inferred [9] for the $\beta$-$\text{Mn}_{20-x}\text{Al}_x$ system, and in [5] the absence of long-range magnetic order was admitted in the $\beta$-$\text{Mn}_{19.8-x}\text{Fe}_x\text{Sn}_{0.2}$ alloys as well. To make a step towards establishing the magnetic state in the $\text{Mn}_{19.8-x}\text{Fe}_x\text{Sn}_{0.2}$ system and relating the modification of its properties to changes in iron concentration, we performed a low-temperature study of the magnetization and magnetic susceptibility of these alloys.

Samples of $\text{Mn}_{19.8-x}\text{Fe}_x\text{Sn}_{0.2}$ alloys with $x = 0.0, 0.5, 1.0, 2.0, 3.0, 4.0, 5.0, 6.0, 6.2$, and 6.4 were synthesized in an induction furnace. For homogenization, the smelted ingots were annealed for 100 hours at 900°C and then quenched with water. X-ray phase analysis...
showed that all the samples are monophase and isosstructural with the $\beta$ modification of manganese. The unit cell parameter monotonically decreases from 6.302 to 6.272 Å as the iron concentration grows from $x = 0$ to $x = 6.4$.

Magnetization and susceptibility in a static magnetic field of up to 10 kOe were measured with a SQUID magnetometer (Quantum Design) in a temperature range from 1.8 to 400 K.

Figure 1 plots the temperature dependences of susceptibility of the Mn$_{19.8-x}$Fe$_x$Sn$_{0.2}$ alloys obtained in a constant field 1000 Oe. It is seen that all the curves are alike in shape resembling the $\lambda$ dependences with well-defined maxima in the temperature range from 15 to 40 K. A similar pattern of peaking dependences $\chi(T)$ was observed in [15] for $\beta$-Mn antiferromagnetic alloys Mn–Fe and Mn–Sn with close composition. To clarify the nature of magnetic ordering at temperatures below the susceptibility peak and to reveal the possible influence of magnetic impurities, we measured magnetization curves for these alloys. The results obtained at a temperature of 4 K are presented in Fig. 2. It is seen that the curves are very much alike and the magnetization is linear with the field strength. The latter circumstance points to the antiferromagnetic nature of the magnetic ordering. The value of magnetization in a fixed magnetic field is almost independent of the iron concentration.

Nevertheless, more accurate measurements revealed that the low-temperature magnetizations of all of the investigated alloys depend on the magnetic prehistory. As an example, Fig. 3 illustrates the behavior of magnetization of the Mn$_{15.8-4}$Fe$_4$Sn$_{0.2}$ samples during the temperature rise from 4 to 30 K; the samples were precooled in the absence of a field and in a field of 10 kOe. It is seen that, at a temperature below 22 K, the sample cooled in a zero field still has a small residual magnetization. As seen at the inset, which shows the region of transition on a magnified ordinate scale, neither of the dependences $M(H)$ has a peak at this temperature. Note that the Neel temperature for this alloy equals to 34 K.