Superantiferromagnetism of cobalt-manganese alloys

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The existence of a superantiferromagnetic state, predicted theoretically by Petrakovskiĭ, Kuz'min, and Aplesnin (Fiz. Tverd. Tela **24**, 3298 (1982) [Sov. Phys. Solid State **24**, 1872 (1982)]), is demonstrated experimentally for the first time using the methods of magnetic neutron diffraction analysis in Co-Mn alloys, whose composition is close to the "zero-scattering matrix."

The Monte Carlo calculations[1] of the magnetic phase diagram of an Ising magnet with random mixing and strongly fluctuating exchange coupling have shown that at certain temperatures and concentrations the topologically infinite antiferromagnetic cluster "decomposes" into stable regions of finite dimensions. Such a magnetic state, which was not previously obtained in calculations performed in the molecular-field approximation,[2,3] was called by Petrakovskiĭ *et al.*[1] a superantiferromagnetic state. The term "superantiferromagnetism" is not new. It was introduced previously by Néel[4] in order to describe the magnetic behavior of an ensemble of finely dispersed antiferromagnetic NiO particles.[5] Its definition as a state of antiferromagnetic clusters in a paramagnetic matrix above the Néel point (T_N) or the temperature of freezing of the spin glass (T_f) differs, however, from the Néel definition and, as far as we know, has still not been confirmed experimentally.

In this paper we present the results of a neutron diffraction analysis of Co-Mn alloys, from which the existence of such a state in real magnetic systems follows unequivocally. The cobalt-manganese alloys first attracted our attention by the unusual behavior of the temperature dependence of the magnetic susceptibility, in which close to the Néel point there were no anomalies.[6,7] As follows from Ref. 1, such a behavior of the susceptibility is also a result of the superantiferromagnetic state.

We studied the elastic coherent scattering of neutrons by disordered alloys of cobalt with a manganese content of 33–52 at. % with the help of a diffractometer, which was inserted into one of the horizontal channels of the IVV-2 reactor. We obtained a monochromatic beam of neutrons ($\lambda = 0.181$ nm) by reflection from the (111) plane of a deformed single crystal of germanium. In this case, the second orders of reflection completely vanished, since the Co-Mn alloys in this region of compositions approached the "zero-scattering matrix" ($b_{Co} = 0.25\times10^{-5}$ nm, $b_{Mn} = -0.36\times10^{-5}$ nm). For this reason, the measured intensity of the scattered neutrons near the (110) reflection had only a magnetic nature, which greatly increased the sensitivity of our method for observing weak coherent reflections.

We have studied the intensity of the scattered neutrons near the (100) and (110) reflections for the alloys studied in the temperature range 4.2–1000 K. For alloys with a manganese content exceeding 43 at. %, a quite narrow coherent reflection (110), indicating an antiferromagnetic ordering of the type γ-FeMn, is observed. At the same time, for the alloys with 35–42 at. % Mn, this reflection, which has a much larger width, indicates that there is magnetic coherent scattering of neutrons by small particles. The concentration dependence of the width of the (110) reflection at half-height at 4.2 K, from which we can see that there are two horizontal sections, which abruptly transform into each other at concentrations of 42–43 at. % Mn, is shown in Fig. 1. An analogous dependence of the width of the reflection is also observed with respect to temperature for alloys with antiferromagnetic long-range order. Up to a certain temperature (T_N), the width of the (110) reflection remains virtually unchanged as its intensity decreases, and then the width increases in a narrow temperature range and the reflection vanishes at another temperature (T_{SAF}). The concentration dependence of T_N and T_{SAF} for the alloys investigated is shown in Fig. 2. This is actually the section of the magnetic phase diagram of Co-Mn alloys in which the region of existence of superantiferromagnetism is located between these two lines.

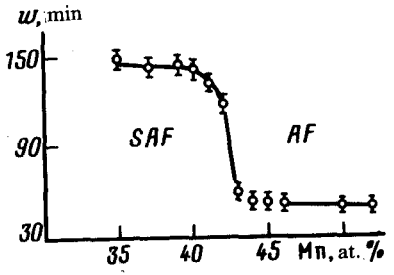

FIG. 1. Concentration dependence of the width of the diffraction reflection (110) at half-height for Co-Mn alloys.

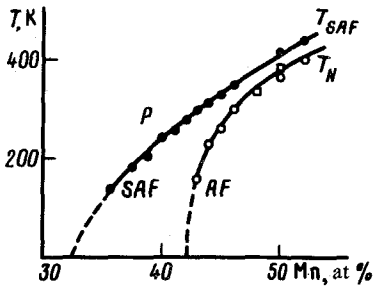

FIG. 2. Section of the magnetic phase diagram of Co-Mn alloys. T_N is the Néel point, T_{SAF} is the temperature of the transition of the superantiferromagnetic state to the paramagnetic state. ○,● are our measurements and □ are the data of Ref. 7.

The dimensions of the final clusters, which appear as regions of antiferromagnetic coherent scattering of neutrons, were estimated using the well-known Selyakov-Sherrer formula, taking into account the experimentally measured width of the (110) line. It turned out that for the compositions 35–42 at. % Mn the size of the antiferromagnetic clusters at 4.2 K varies from 8 to 11.5 nm. Antiferromagnetic regions with approximately the same size also exist above the Néel temperature of alloys with a manganese content of 43–52 at. %.

Thus the state of the antiferromagnet, which is fragmented into finely dispersed sections ~ 10 nm in size in a paramagnetic matrix, corresponds exactly to the definition of superantiferromagnetism given in Ref. 1. This magnetic state of Co-Mn alloys is a result of a competing exchange interaction of the type $J_{CoCo} > 0$, $J_{CoMn} < 0$ and $J_{MnMn} < 0$, which we will discuss in greater detail in subsequent papers.

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