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Diffuse nuclear and magnetic neutron scattering in quenched $\mathbf{Co_{1-x}Mn_x}$ alloys

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Abstract. Diffuse neutron scattering measurements were performed on quenched $Co_{1-x}Mn_x$ alloys with $x = 0.31$, 0.36, 0.39, 0.43 and 0.5, which belong to the transition region between ferromagnetism and antiferromagnetism. The nuclear and magnetic cross sections were separated, and Cowley short-range order atomic parameters determined. It was found that the superantiferromagnetic behaviour of these alloys occurred owing to the existence of the antiferromagnetic clusters of the γ-FeMn type caused by short-range order regions of the CuAu type.

1. Introduction

The $Co_{1-x}Mn_x$ alloys have a peculiar place among 3d transition binary alloys with a FCC lattice. They provide a chance to follow the concentration transition from ferromagnetism to antiferromagnetism within the framework of one type of solid solution. The magnetic properties of these alloys in ferromagnetic and antiferromagnetic regions have been investigated in earlier work [1–7]. A more detailed study of $Co_{1-x}Mn_x$ alloy magnetic states, including magnetic and neutron diffraction measurements, has been carried out by Menshikov and co-workers [8, 9]. As a result of these investigations a magnetic phase diagram was drawn where the regions of long-range ferromagnetic and antiferromagnetic order, as well as the transition region $0.25 < x < 0.42$, were determined. It has been observed that the $Co_{1-x}Mn_x$ alloys in the latter region have cluster magnetism properties. This conclusion has been made from the discovery of a broad magnetic (110) reflection in the alloys with $x = 0.36$–0.42 because of the existence of antiferromagnetic clusters of approximately 100 Å sizes. It was surprising because in similar quasi-binary alloys, e.g. γ-FeNiMn [10], a spin-glass state took place in the transition region.

Unfortunately, in our previous magnetic neutron scattering study [8, 9] the intensity of the (110) reflection has been measured against the background of diffuse nuclear scattering. Therefore there was no possibility of making a conclusion on the diffuse nuclear scattering. That disadvantage has been overcome in the recent work by Wildes *et al* [11]. These workers separated the nuclear diffuse scattering component in the alloys with $x = 0.32$ and $x = 0.37$ by neutron polarization analysis. They discovered also that there was no magnetic diffuse scattering (within their error) for these alloys in the transferred moment region $q = 0.2$–2.5 Å^{-1}. However, the transferred moment region noticed did not exceed the range of existence of the (110) reflection, where the magnetic diffuse scattering was measured in our work [8, 9]. Recently Cable and Tsunoda [12, 13] have measured very carefully neutron diffuse scattering in single crystals of $Co_{1-x}Mn_x$ alloys, which belong to transition region ($x = 0.25$, 0.30, 0.35 and 0.40). They have observed diffuse magnetic scattering for these alloys indicative of antiferromagnetic short-range order with a very small manganese magnetic moment of about $1\mu_B$.

The purpose of the present study was to elucidate the role of short-range atomic order effects in the formation of superantiferromagnetism (or short-range antiferromagnetic order) in the $Co_{1-x}Mn_x$ alloys. For that we chose five most typical alloys with $x = 0.31$, 0.36, 0.39, 0.43 and 0.50 from a large number of alloys investigated earlier [9]. All the enumerated alloys were in the condition close to the 'zero matrix' for an average nuclear scattering amplitude. It allowed us to distinguish between the nuclear and magnetic components of diffuse neutron scattering at large transferred moments from $q = 0.3$ up to 3.5 $Å^{-1}$ with great accuracy.

Diffuse scattering of non-polarized neutrons was measured with a diffractometer installed at the IVV-2M reactor (Ekaterinburg, Russia). A monochromatic neutron beam of wavelength $\lambda = 1.81$ Å was obtained by reflection from two crystals (germanium and pyrographite). It allowed us to avoid $\lambda/2$ contamination. Neutron diffraction measurements have been performed at liquid-helium temperature, room temperature and 500 K. As a result of these measurements, magnetic scattering and nuclear diffuse scattering were separated and Cowley short-range order atomic parameters were determined.

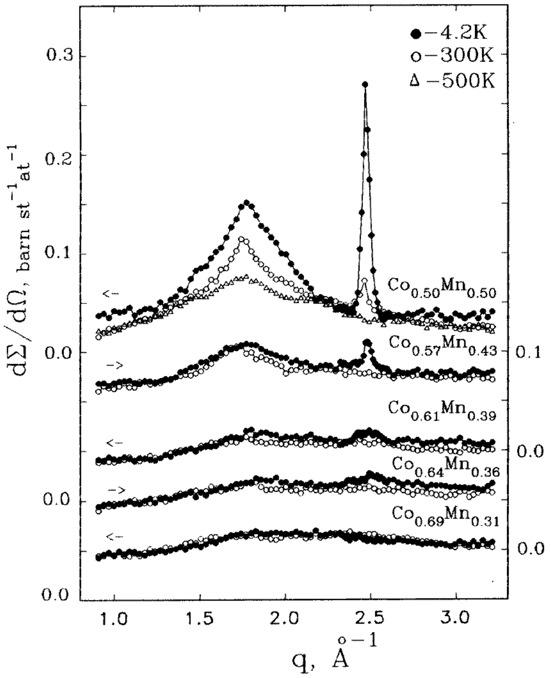

Figure 1. The experimental diffuse neutron scattering patterns obtained for $Co_{1-x}Mn_x$ alloys at 4.2 K (○), 300 K (●) and 500 K (△).

2. Experimental results

Experimentally measured angle dependences of diffuse neutron scattering are presented in figure 1 for all the alloys investigated at 4.2 and 300 K and also for a sample of $Co_{0.5}Mn_{0.5}$

at 500 K. One can see that there is no difference between the diffuse scattering patterns obtained at 4.2 and 300 K for the alloy with $x = 0.3$. However, this difference becomes apparent in the transferred moments region close to the (110) magnetic reflection with increasing x and, for the alloy with $x = 0.39$, a weak coherent (110) reflection takes place. Its intensity becomes rather high for the alloy with $x = 0.5$. Also, a variation between the two neutron diffraction patterns in the range of (100) reflection exists.

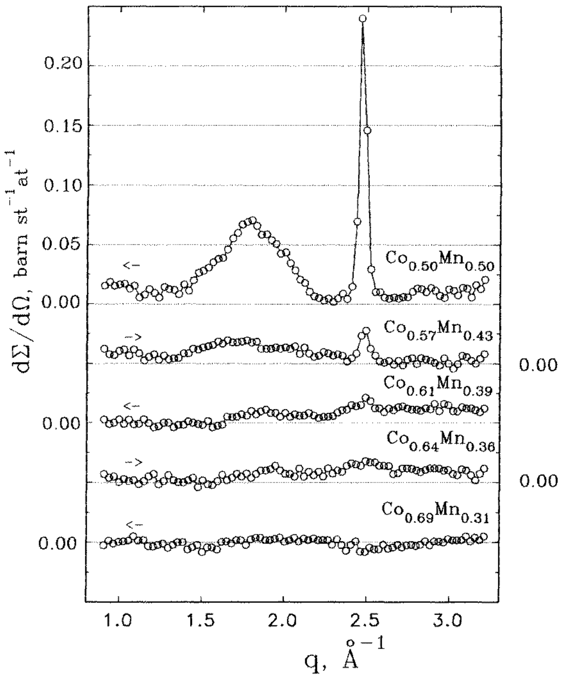

Figure 2. The magnetic diffuse neutron scattering for $Co_{1-x}Mn_x$ alloys.

Figure 2 shows the magnetic diffuse scattering component in $Co_{1-x}Mn_x$ alloys obtained as a difference between the two neutron patterns. This magnetic scattering is caused by the existence of short-range antiferromagnetic order in the alloys with $x < 0.43$ and long-range order in the alloys with $x \geqslant 0.43$. The antiferromagnetic structure of these alloys is described by the wavevector $\boldsymbol{k} = (2\pi/a)(0, 0, 1)$. The existence of incoherent magnetic scattering near the (100) reflection proves the manifestation of uncorrelated x- and y-projections of a full magnetic moment in the magnetic structure of the γ-FeMn type. The fit to the experimental nuclear incoherent scattering of the atomic short-range order function is shown in figure 3 as an example for $Co_{69}Mn_{31}$ alloys.

3. Discussion

According to the magnetic phase diagram in figure 4, taken from our work [6], the alloys with $x < 0.43$ are paramagnets at room temperature while the alloy $Co_{0.5}Mn_{0.5}$ is a paramagnet at $T = 500$ K.

Therefore, the neutron diffraction patterns obtained at these temperatures completely correspond to nuclear neutron scattering with the accuracy of paramagnetic scattering. The latter vanishes in our case, because of the small average magnetic moments in these

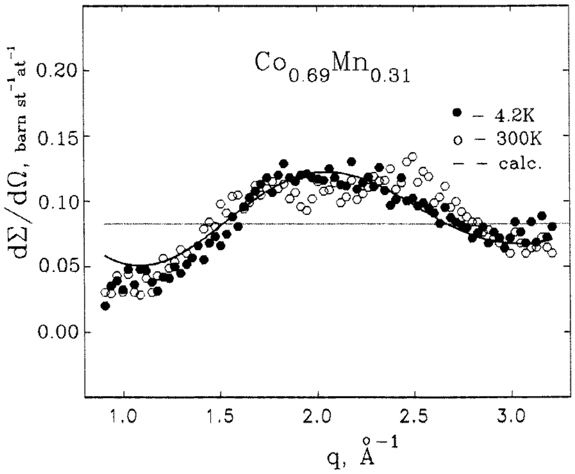

Figure 3. The fit to the experimental nuclear diffuse scattering curve of the atomic short-range order function for $Co_{69}Mn_{31}$ alloys.

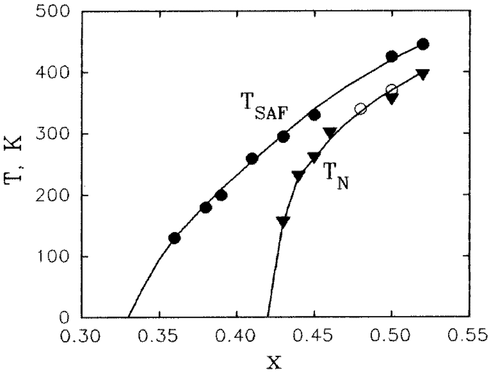

Figure 4. Magnetic phase diagram of $Co_{1-x}Mn_x$ alloys in the region of superantiferromagnetic-to-antiferromagnetic transition.

alloys [12, 13]. The existence of the diffuse maxima in the place of the (100) and (110) superstructural reflections proves the presence of the short-range atomic order effects. To calculate the parameters of this order we use the differential cross section of neutron scattering as follows [14]:

$$\frac{d\sigma}{d\Omega} = x(1-x)(b_{Co} - b_{Mn})^2\left(1 + \sum_i Z_i a_i \frac{\sin(q_i R_i)}{q_i R_i}\right).$$

Here $b_{Co} = 0.253 \times 10^{-12}$ cm and $b_{Mn} = -0.373 \times 10^{-12}$ cm are the nuclear scattering amplitudes, Z_i is the number of atoms in the ith sphere. α_i is the Cowley short-range order parameter. $\alpha_i = \varepsilon^i_{MnMn}/x(1-x)$ where $\varepsilon^i_{MnMn} = xp^i_{MnMn} - x^2$ is the atomic correlation parameter. ε^i_{MnMn} and α_i are negative if the probability of finding an Mn atom in the corresponding coordination sphere p^i_{MnMn} is less than the average Mn concentration, $x(p^i_{MnMn} < x)$, and they are positive if $p^i_{MnMn} > x$. The first situation is present for short-range order, and the second for short-range segregation; in a completely disordered alloy, $p^i_{MnMn} = x$.

The Cowley parameters calculated by the least-squares method are presented in table 1. It is seen, that the α-values for the first coordination sphere are negative, and the absolute values of these parameters increase with increasing Mn content.

Table 1. Short-range (α_i) and long-range (η_*) order parameters in $\mathrm{Co}_{1-x}\mathrm{Mn}_x$ alloys. $\Delta\alpha = \pm 0.001$; $\Delta\eta_* = \pm 0.01$.

x	α_1	α_2	α_3	$\varepsilon^{(1)}_{MnMn}$	η_*
0.31	−0.029	0.127	0.006	−0.006	0.30
0.36	−0.037	0.120	0.010	−0.008	0.33
0.39	−0.043	0.110	0.023	−0.010	0.36
0.43	−0.048	0.076	0.040	−0.012	0.38
0.50	−0.055	0.062	0.016	−0.014	0.40

The short-range order parameters corresponding to the atomic correlation $\varepsilon^{(1)}_{MnMn} = \alpha_1 x(1-x)$ may be renormalized to the long-range order parameter η_*. In the case when there is no coexistence of short-range and long-range atomic orders the probability that the Mn atom is next to the Mn atom in solid solution ordered as a type of CuAu may be found as follows [14]:

$$p^i_{MnMn} = x - \frac{\gamma^2\eta_*^2}{3x} + \frac{\varepsilon^{(1)}_{MnMn}}{x}$$

where $\gamma = x(1-\nu)/\nu$ for $x \leqslant \nu = 0.5$. We propose that $|\varepsilon^{(1)}_{MnMn}/x|$ cannot be more than $\gamma^2\eta_*^2/3x$. Therefore $\eta_*^2 = 3\varepsilon^{(1)}_{MnMn}/x^2$. From table 1, the value of the long-range order parameter $\eta_* \simeq 0.3$–0.4 exists for all the alloys investigated. However, the value of η_* is not sufficiently high for the treatment of tetragonality in these alloys. Therefore, this testifies to the short-range atomic order of the CuAu type responsible for the superantiferromagnetism in the quenched $\mathrm{Co}_{1-x}\mathrm{Mn}_x$ alloys in which the antiferromagnetic short-range order structure is described by the type of cubic γ-FeMn alloy. As usual it is characterized by incoherent neutron scattering in the place of (100) reflection. This is related to the existence of uncorrelated x- and y-projections of full magnetic moment slowly relaxing around a chosen axis.

4. Conclusion

The investigations proved the existence of the short-range atomic order of the CuAu type in the the $\mathrm{Co}_{1-x}\mathrm{Mn}_x$ alloys of composition close to equiatomic. It causes antiferromagnetic cluster formations of the γ-FeMn type, observed in alloys with Mn content in the range $0.31 < x < 0.43$. It is obvious that the centres of such magnetic clusters (superantiferromagnetism) are the short-range atomic order regions, the sizes of which are less than those of magnetic regions since the nuclear diffuse scattering occupies a broad angle range.

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