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Neutron Diffraction Investigation of Order-Disorder in the Alloys "Ferrum-Nickel" and "Ferrum-Cobalt"

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The large single crystals of Fe-Ni alloys belonging to the permalloy class are investigated. Among them are binary alloys of Fe and Ni and ternary alloys of Fe and Ni with Mo, Cr and Cu which are almost the same as of industrial marks. The new results are obtained for the equilibrium phase of alloys on the basis of thermodynamics of order-disorder transformation. The dependence of magnetic anisotropy constant on the degree of long range order in Ni_3Fe alloy is given.

The atomic order-disorder phenomena in Fe-Co alloys which have b.c.c. crystal lattices are investigated using polycrystal sample. The existence of the equiatomic superlattice over a wide range in Fe-Co system was discovered: approximately from 25% Co to the boundary of $\alpha \rightleftarrows \gamma$ transformation (~75% Co).

It is concluded that the energy of order-disorder transformation of Fe-Ni and Fe-Co alloys is in general due to $3d$-exchanged interaction.

1. Introduction

The solid solutions of Fe-Ni and Fe-Co which show the atomic order-disorder transformation have specially interesting points among the magnetic alloys. In ferrum-nickel and ferrum-cobalt series one could get both high permeability alloys (permalloy, supermalloy, hiperco, permendur) and hard magnetic alloys (vicalloy, alnico, magnico and others). However, the influence of the degree of atomic order on the magnetic properties of the solid solutions as well as the nature of the order-disorder transformation in these alloys have not been studied enough because of the difficulties of the definition of the order degree for these alloys. Fe, Co and Ni which are practically indistinguishable by the X-ray analysis become essentially different atoms for the neutron diffraction on account of the difference in the scattering powers (see Table I). This fact is clearly shown in the investigations of the superstructures of FeCo[1] and Ni_3Fe[2]. In the present work further studies of above mentioned systems were made.

Since only the magnetic part of the neutron scattering in the permalloys which is only about 1/10 of the nuclear contribution can give the necessary information about the order degree, Lowde's method was applied[3]. In this case the (010) magnetic superlattice line which intensity is about 3/4 of the inten-

Table I.

Elements and superstructures	X-rays ($\sin\theta/\lambda=0.5$Å^{-1})		Neutrons			
			Nuclear scattering		Magnetic scattering ($\sin\theta/\lambda=0.5$Å^{-1})	
	Amplitude $f\times10^{-12}$cm.	$F^2_{sup.}/F^2_{fund.}$	Amplitude $f\times10^{-12}$cm.	$F^2_{sup.}/F^2_{fund.}$	Amplitude $f\times10^{-12}$cm.	$F^2_{sup.}/F^2_{fund.}$
28. Ni	3.58		1.03		~0.05	
Ni_3Fe		0.02		0.02		~0.43
26. Fe	3.27		0.96		~0.20	
FeCo		0.0004		0.3		~0.
27. Co	3.41		0.28		~0.20	

sity expected for the perfectly ordered Ni_3Fe is observed on the background of the $\lambda/2$ of fundamental nuclear reflexion (020). For the Ni_3Fe alloy annealed 600 hrs at temperatures between 600→300°C the long range order parameter σ was given to be equal to 0.75 in accord with the saturation of the physical properties. The identical heat treatment was applied to all other permalloy specimens.

The polycrystal specimens of the Fe-Co alloys were investigated by the neutron Debye-Scherrer's method. The same heat treatments were made for all Fe-Co alloy specimens: the annealing was carried out in the interval of 800→400°C within one week.

2. Results

The chemical composition of the investigated Fe-Ni alloys and the obtained σ values are given in Table II. The neutron diffraction pattern of the Ni_3Fe alloy is shown in Fig. 1. In Fig. 2 the dependence of the parameter σ on the temperature of the isothermal annealing in this alloy is shown. The measurements of the neutron diffraction patterns were carried out at room temperature after quenching the specimens of the preliminary ordered alloy at respective temperatures. The magnetic anisotropy constant K at liquid nitrogen temperature was measured at the same time for each specimen. The obtained dependence $K=K(\sigma)$ is shown in Fig. 3. The measured critical point of the order-disorder transition is nearly equal or higher than 530°C for Ni_3Fe alloy while heating. The temperature hysteresis of the transformation at cooling was found as approximately 10°.

Table II.

No. of specimens	Chemical composition (at. %%)			Parameter σ
	Ni	Fe	Third element	
1	49.3	50.7	—	0.
2	69.7	30.3	—	0.9
3	75.5	24.5	—	0.75
4	81.6	18.4	—	0.
5	78.8	18.9	2.3%Mo	0.
6	79.4	17.2	3.4%Mo	0.
7	73.4	23.6	3.0%Cr	0.75
8	74.6	21.7	3.7%Cu	0.3

Linear sizes of the long range order domains were estimated to be about 30Å from the width of the superlattice line (010) and the Selakov-Scherrer's formula.

Both the neutron diffraction pattern of the ordered FeCo alloy shown in Fig. 4 and those of other alloys of the Fe-Co systems show only the regular variations of the fundamental reflexion and background intensities. Actually, chemical composition varied continuously in these Fe-Co systems. The visible expansion of the superlattice has not been observed. The concentration dependence of the para-

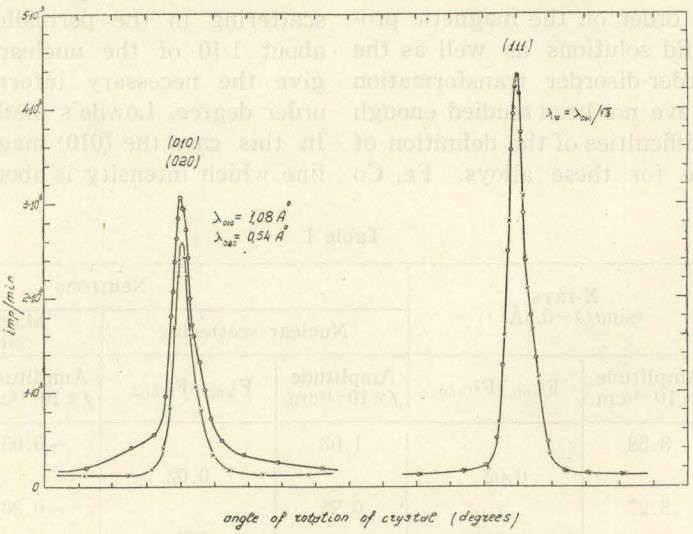

Fig. 1. Curves of intensity of fundamental reflexion (111) and reflexion (010) with all harmonics for order (○) and disorder (×) specimen states.

meter σ at room temperature is shown in Fig. 5. The preliminary data for the temperature dependence of σ for the FeCo alloy[4] are given in Fig. 6.

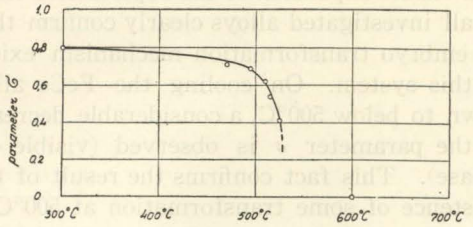

Fig. 2.

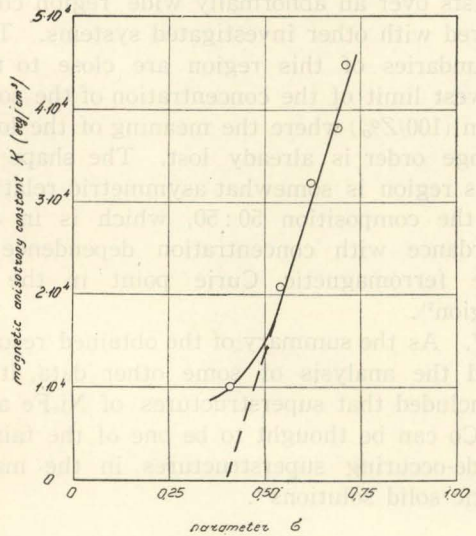

Fig. 3.

3. Summary

1. The sharp asymmetry of the concentration region of Ni_3Fe superstructure was obtained in relation to the stoichiometric composition. This is due to the asymmetry of the well known diagrams, namely the composition-variation of the properties of the binary Ni-Fe alloys under different heat treatments. The highest value of σ in comparison with the parameter σ for the Ni_3Fe alloy was obtained for 70% Ni alloy. It proves that the states of the investigated alloys are not in the thermal equilibrium and that order-disorder energy in these alloys is apparently different from the one obtained. These facts confirm the Smoluchowski's hypothesis about the essential share of the $3d$-interaction energy of the order-disorder energy in the Fe-Ni alloys and the close connection of the concentration dependence of the ferromagnetic Curie point upon the shape of the alloy equilibrium diagram in the order-disorder region[5].

2. For the equiatomic alloy of NiFe the laminated ferromagnetic superstructure of the CuAu or CuPt types was not obtained. The known influence of the heat treatment of alloys on its physical constants and properties can be explained by the order-disorder processes of the positive or negative short range order types.

3. The superlattice reflexions in the Mo-permalloy and supermalloy were not observed as well. In the Cu-added 75%-permalloy the long range order degree is small. In the Cr-added 74%-permalloy, however, it is found

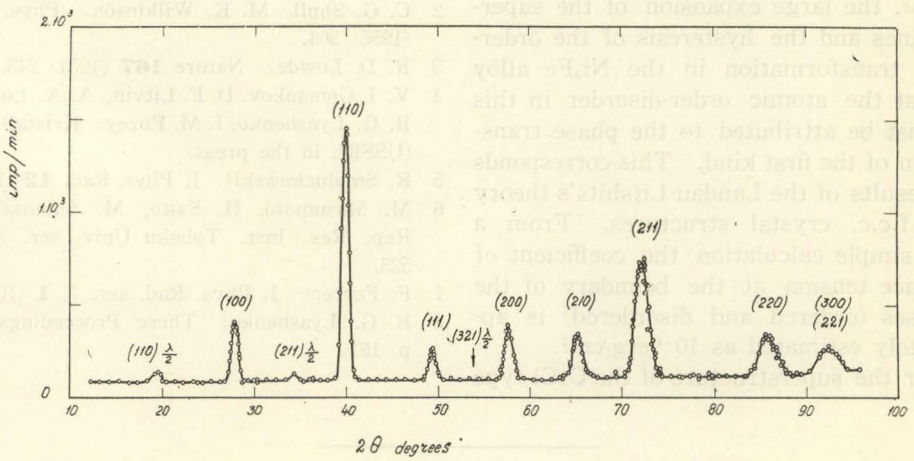

Fig. 4.

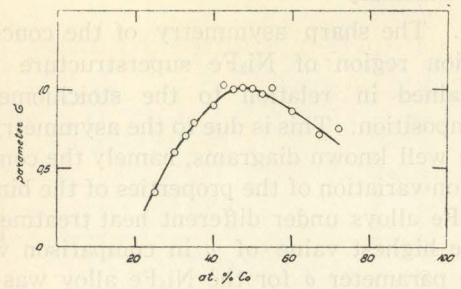

Fig. 5.

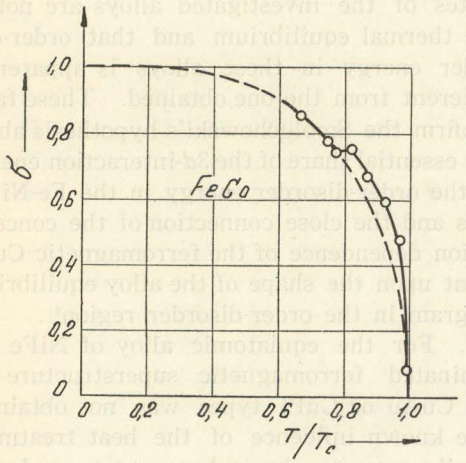

Fig. 6.

that the superstructure is highly developed. As the magnetic induction of these alloys decreases abruptly by adding Cr, one can expect that the Cr atoms interact antiferromagnetically with the neighbours and take active part in the creation of the triple superstructure of $Ni_3(Fe, Cr)$.

4. The shape of the temperature dependence of σ, the large expansion of the superlattice lines and the hysteresis of the order-disorder transformation in the Ni_3Fe alloy show that the atomic order-disorder in this alloy must be attributed to the phase transformation of the first kind. This corresponds to the results of the Landau-Lifshits's theory on the f.c.c. crystal structures. From a slightly simple calculation the coefficient of the surface tension at the boundary of the two phases (ordered and disordered) is approximately estimated as 10^{-1} erg/cm^2.

5. For the superstructure of the CsCl type in α-solid solution of Fe and Co, Landau-Lifshits's veto on the order-disorder phase transition of the second kind does not exist. The shape of the temperature dependence of σ for the equiatomic alloy and also the absence of a visible expansion of the superlattice lines in all investigated alloys clearly confirm that no embryo transformation mechanism exists in this system. On cooling the FeCo alloy down to below 500°C, a considerable decrease of the parameter σ is observed (visible decrease). This fact confirms the result of the existence of some transformation at 500°C[6].

6. By a relatively rapid cooling of the Fe-Co alloys from high temperatures, it was found that the superstructure of this alloy exists over an abnormally wide region compared with other investigated systems. The boundaries of this region are close to the lowest limit of the concentration of the solution $(100/Z\%)$ where the meaning of the long range order is already lost. The shape of this region is somewhat asymmetric relative to the composition 50 : 50, which is in accordance with concentration dependence of the ferromagnetic Curie point in the γ-region[7].

7. As the summary of the obtained results and the analysis of some other data, it is concluded that superstructures of Ni_3Fe and FeCo can be thought to be one of the fairly wide-occuring superstructures in the magnetic solid solutions[8].

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