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OBSERVATION OF A RELAXATION PHENOMENON DURING AGEING OF Fe-Ni-C MARTENSITE BY MEANS OF SMALL ANGLE NEUTRON SCATTERING

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Introduction

The Fe-Ni-C martensite with a high content of nickel (over about 18 wt%) is characterized by abnormally high tetragonality (1,2). The nature of high tetragonality of virgin martensite as well as the phenomena responsible for its reduction at early stages of ageing remain so far an object for discussion. Among the models proposed, one can point out redistribution of carbon atoms (3,4) and the break of coherency between the freshly formed martensite and austenite (5,6). The technique of small angle scattering of neutrons was applied to get information about the phenomena occurring at the early stage of ageing of the Fe-25Ni-0.7C (wt%) martensite formed during cooling to liquid nitrogen temperature. The scattering of polarized neutrons was measured at room temperature. A relaxation caused by the change of the magnetic structure was observed. It is assumed that diffusion of carbon atoms changes the nearest neighbourhood of iron and nickel atoms at an early stage of ageing and leads to changes in the inhomogeneous magnetic structure of the solid solution.

Material and techniques

The alloy Fe - 25 wt% Ni - 0.7 wt% C was chosen for the present study to get high-tetragonal virgin martensite. Four sample plates with dimensions 45 x 10 x 15 mm^3 were used. After solution treatment for 3 hours at 1100 °C the samples were quenched in water. The alloy was wholly austenitic at room temperature. Martensite was formed during quenching in liquid nitrogen, the M_s temperature being 140 K. Before the martensitic transformation the magnetic ordering of austenite was observed by means of magnetic susceptibility measurements. The Curie temperature was 190 K.

The momentum and time dependences of the intensity of the polarized neutron small angle scattering were studied at 300K before and after the martensitic transformation. The measurements were performed using multidetector equipment for polarized neutrons as decribed elsewhere (7). The equipment parameters were the following: the average neutron wavelength $\lambda = 1$ nm ($\Delta\lambda/\lambda = 30\%$) and the range of momenta transferred $3 \cdot 10^{-4} < q < 5 \cdot 10^{-2}$ nm^{-1}. The resolution of q approximated by the Gaussian function was $5 \cdot 10^{-4}$ nm^{-1}.

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The measurements on the first sample were carried out for 5 hours before quenching in liquid nitrogen and after quenching for 16 hours without an external magnetic field. After that the sample was annealed at T=333 K and at T=373 K in order to check if further relaxation had occurred at these temperatures. The second sample was studied at the same time and temperature sequence in the magnetic field H = 0.3 T. Before annealing the H-dependence of the scattering intensity was measured in the range of H = 0 - 0.3 T. The magnetic field was applied parallel to the scattering vector, because in this geometry elastic magnetic scattering is suppressed due to the magnetic saturation.

The intensity of the scattering was determined as a difference between the measured intensity and the background as observed on the austenitic sample before cooling to liquid nitrogen temperature.

Experimental

Fig.1 shows a typical time dependence of the neutron scattering intensity, normalized to the beginning of the relaxation observations. In the lower part of the figure two points are given for scattering at two fields (0 and 0.3 T) after annealing at 333 K and at 373 K. The scattering intensities for these two annealing temperatures are the same within the limits of error.

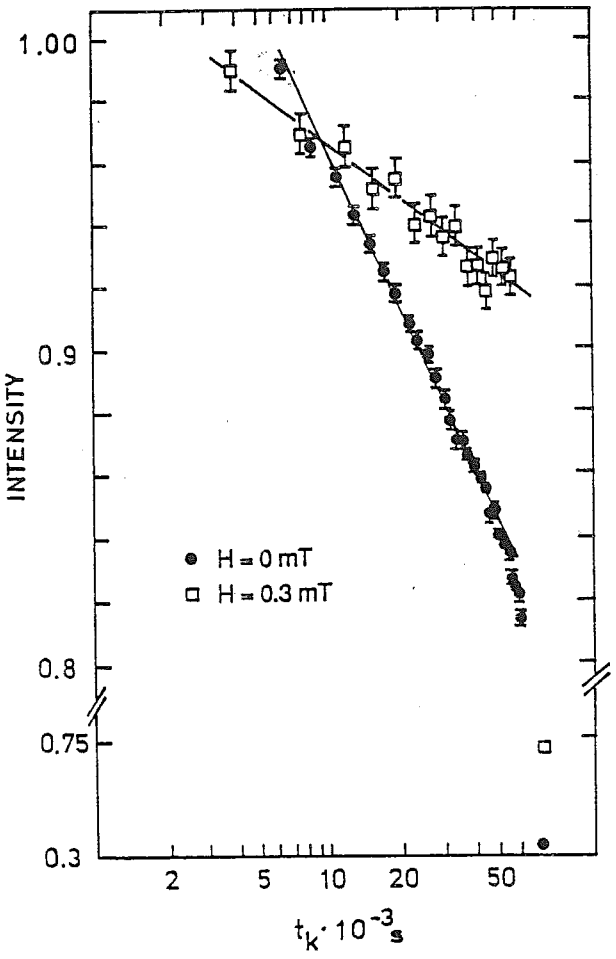

FIG. 1. Time dependence of the intensity of neutrons scattered with the momentum transferred q = 0.1 nm^{-1} for the alloy Fe-25Ni-0.7C (wt%) after quenching in liquid nitrogen; measured at 293 K. Scattering levels after heating to 333 K and 373 K are shown in the lower part of the figure.

The time dependence of the intensity I(q,t) was described by a logarithmic law

$$I(q,t) = I_0(q) - S \log(t/t_0),$$

where t_0 is the characterictic time of the measurement (t_0=3600 s) and S is the aftereffect coefficient. The dependence of the aftereffect coefficient S on the momentum transferred is shown in Fig. 2 for the two types of measurements.

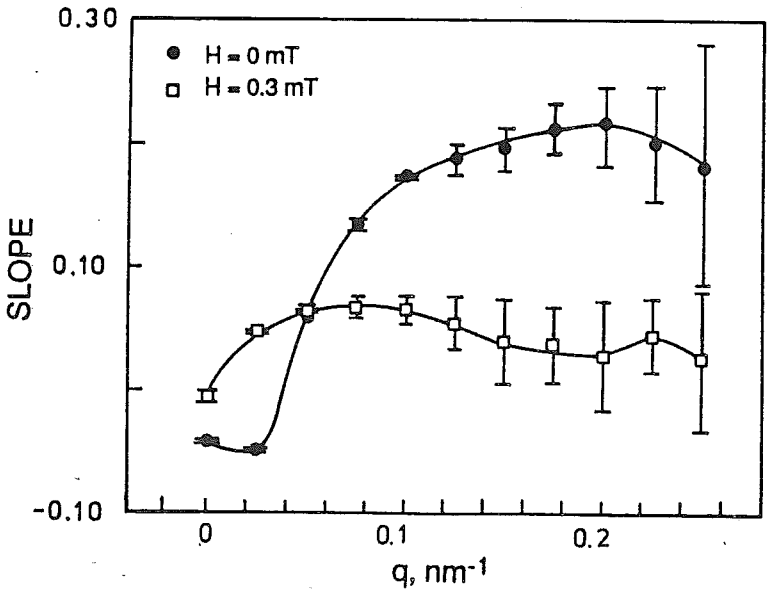

FIG. 2. The dependence of the aftereffect coefficient on the momentum transferred

The small angle scattering measured is characterized by the following peculiarities:

a) Strong scattering is observed throughout the whole range of momenta transferred.

b) Complete depolarization of neutrons exists in magnetic fields up to 0.3 T, which means the absence of magnetic saturation.

c) The intensity of scattering strongly depends on the applied field value (Fig. 1), for example it decreases by over a decade at the field of H = 0.3 T as compared to H = 0. It means that the scattering observed has a magnetic nature i.e. it is caused by magnetic inhomogeneities corresponding probably to the structural peculiarities of the alloy.

d) The rise of the relaxation rate is accompanied by an increase in the momentum transferred (Fig. 2) i.e. with a decrease in the scale of the space inhomogeneities. The inverse sign of the coefficient S at q = 0 (the increase of the scattering intensity with time for small momenta transferred and at the same time the bigger scattering for large q) shows the change of the inhomogeneous structure, with time, towards a decrease in the contribution of the small regions to the scattering intensity.

Discussion

The logarithmic dependence of the relaxation time is a typical property of strongly degenerated systems with a continuous relaxation spectrum as observed for example in spin glasses. If we assume that a system is in a state of equilibrium after annealing at 333 K or 373 K and the relaxation phenomenon observed leads to this equilibrium with the rate S in the logarithmic scale, it is possible to evaluate the characteristic relaxation time, as it is performed for spin glasses (8,9):

$$t_{max} = t_I \cdot 10^{\Delta I/S},$$

where ΔI is the difference of intensities for a given value of q between the annealing and the beginning of relaxation after quenching and t_I is a typical time of measurements (t_I is the order of t_0).

The preliminary analysis shows that the characteristic relaxation time t_{max} is approximately the same for the relaxation phenomena at H = 0 and at H = 0.3 T. There is a momentum dependence of t_{max} that is t_{max} is nearly 1.5 times bigger in the scales of $q < 0.1$ nm^{-1} as compared to the scales of $q \geq 0.1$ nm^{-1}, a typical order of $\Delta I/S$ being approximately 5.

The time dependence of the relaxation will be discussed in the following. The iron-nickel alloys are characterized by an inhomogeneous magnetic structure (10-12). A number of theoretical models have been proposed, among them the one by Sidorov and Doroshenko (13) is one of the most typical. According to these authors a non-collinear magnetic structure exists as a result of different (ferromagnetic and antiferromagnetic) interactions between atoms of different kinds. It means that atomic magnetic moments are oriented at different angles to the direction of spontaneous magnetization depending on the nearest neighbourhood, Z projections of atomic magnetic moments being only collinear.

The short range atomic ordering is observed in Fe-Ni alloys (14,15) and interstitial elements are shown to assist the phenomenon of ordering (16,17). According to Mössbauer data (18) the dissolution of carbon atoms in Fe-Ni austenite promotes an inhomogeneous short range atomic ordering with the decomposition of the solid solution into submicroregions enriched or depleted in nickel and correspondingly depleted of or enriched with carbon.

The inhomogeneous distribution of nickel and carbon atoms in austenite must be inherited by martensite during the diffusionless martensitic transformation. It means that virgin martensite is characterized by the inhomogeneous distribution of nickel and carbon atoms which causes its inhomogeneous magnetic structure.

On the basis of the considerations discussed above we can interpret the data obtained in the following way: the change of magnetic structure during ageing of the Fe-25Ni-0.7C martensite can be caused only by a redistribution of carbon atoms, because the crystal lattice imperfections can not change either atomic magnetic moments or their Z projections, and nickel atoms are essentially immobile in the characteristic time of measurements performed. However, carbon atoms begin to move below room temperature and form a modulated structure as a result of spinodal decomposition (19). The migration of carbon atoms changes the nearest neighbourhood of iron and nickel atoms, which is accompanied by the change of atomic magnetic moments, and leads to the change of the neutron scattering caused by magnetic inhomogeneities.

The hypothesis proposed agrees with the above mentioned peculiarities of the neutron scattering. The rate of relaxation is highest in the smallest regions of inhomogeneity where the migration of carbon atoms quickly changes the average neighbourhood. The characteristic relaxation time t_{max} is biggest for the largest scales of inhomogeneity, because a long range migration of carbon atoms is required to achieve the equilibrium state.

Summary

The changes in the small angle neutron scattering are observed during the early stages of ageing of the freshly formed Fe-Ni-C martensite. These changes are attributed to the migration of carbon atoms. The further application of the small angle neutron scattering techniques can provide the information about the scales of the inhomogeneous distribution of carbon atoms in martensite and about some thermodynamical parameters of carbon atom migration.

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