Magnetic inhomogeneities in temperature dynamics of spin structure of the ordered Fe\textsubscript{100-x}Al\textsubscript{x} (25<x<35 at.%) alloys

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Abstract. The results of temperature and in-field Mössbauer and magnetometric studies of the ordered Fe\textsubscript{100-x}Al\textsubscript{x} (25<x<35 at.%) alloys are discussed. Interpretations of observed behaviour of magnetic characteristics versus temperature in terms of the model of collinear magnet with magnetic inhomogeneities on a nanometer scale are proposed.

1. Introduction
Concepts of the origin of the unusual magnetic properties of the ordered Fe\textsubscript{100-x}Al\textsubscript{x} (25<x\leq35 at.%) alloys are varied [1-4]. In some concepts [1,5] it was supposed that the magnetic inhomogeneities - two types local regions: surroundings of the type “Fe\textsubscript{3}Al” and surroundings of the type “FeAl” - govern their magnetic properties. Neutron scattering studies [6] concluded that these alloys comprised ferromagnetic Fe\textsubscript{3}Al-like regions consisting of Fe atoms with $k_{Fe}\geq4$ nearest neighbouring Fe atoms, and the regions consisting of Fe atoms with $k_{Fe}<4$. With increasing the temperature the regions (or clusters) arranged by Fe atoms with $k_{Fe}<4$ exhibit a superparamagnetic-like behaviour. Recent neutron scattering data of B2-ordered Fe-Al alloys [7] proved out the existence of spatial correlation of magnetic moments with a coherence length $\approx5$ nm, that was interpreted in terms of incommensurate spin density waves. But, the conditions of spin density wave origin in these partially disordered alloys remained yet unclear. In \textit{ab-initio} calculations of the local magnetic moments of Fe-Al alloys (from 29 to 44 at.% Al) [8] three different solutions were found: two collinear states (a ferromagnetic one and a state with Fe magnetic moments oriented parallel and antiparallel to magnetization) and a spin-spiral wave. In this work we propose the explanation of the observed temperature behaviour of magnetic characteristics within the simple model of Fe atom local magnetic moment [9], with its magnitude and direction being dependent on the number of Al atoms in its nearest neighbourhood.

2. Experimental
Ingots of Fe-Al alloys with Al concentration 25<x<35 at.% were arc melted from high-purity components in purified He and homogenized at 1100C for 8h in purified Ar. The powders were...
subjected to a heat treatment to obtain B2 or D03 superstructure. The preparation and X-ray diffraction analysis are given in [10] in detail. A chemical analysis was performed using a Spectraflame-Modula D atomic emission spectrometer with an inductively coupled plasma and an MC7201 secondary-ion mass spectrometer. The content of Al in the alloys was 26.5, 30.0, 32.6, and 34.1 at. % to an accuracy of ± 0.3 at. %. Magnetic and Mössbauer measurements of bcc samples with various types of order: Fe73.5Al26.5 (D03), Fe70Al30 (D03), Fe67.4Al32.6 (B2), and Fe65.9Al34.1 (B2) were performed. The magnetic measurements were carried out using an MPMS-XL-5 (Quantum Design) SQUID magnetometer in external magnetic fields $H_{\text{ext}}$ to 50 kOe at 5 - 700 K. The $^{57}$Fe Mössbauer spectra were taken using a spectrometer with a $^{57}$Co in the Cr matrix at 6 - 700 K. To calculate the distributions of the hyperfine magnetic field (HFMF) $P(H_{hf})$, the regularization method was used. The interval of hyperfine fields from 0 to 30 kOe, where the $P(H_{hf})$ distribution corresponds to the spectrum part without magnetic splitting, was taken into account in the calculations of the average HFMF $H_{\text{avg}}$ by putting $H_{hf} = 0$ for the $P(H_{hf})$ in this interval.

3. Results and discussion

3.1. Magnetic measurements

Only for the Fe73.5Al26.5 alloy the magnetization curves and ZFC and FC cycles are close to canonical ferromagnetic systems. The magnetization of the rest alloys under study does not reach saturation in an external field $H_{\text{ext}}$ of up to 50 kOe. Magnetic-hysteresis loops of all alloys are unshifted and symmetric. In the ZFC and FC curves for all the alloys in low $H_{\text{ext}}$ a clearly pronounced thermomagnetic hysteresis is observed at $T \approx 40$ K. Detailed comparison of the ZFC and FC curves values with the values of the magnetization in magnetic hysteresis loops at corresponding $H_{\text{ext}}$ and temperature showed that the true reason of the ZFC and FC curves hysteresis is field-related hysteresis rather than thermomagnetic one [9]. The absence of hysteresis in $H_{\text{ext}}$ exceeding 8-10 kOe, i.e., at those values of $H_{\text{ext}}$ when the dependence of the magnetization on the field has a one-to-one relation (figure 1), sustains such an understanding of the ZFC and FC experiment results. One more peculiarity of $\sigma(T)$ in FC and ZFC cycles is pronounced maxima, with position being dependent on Al concentration in an alloy. The temperature dependence of AC susceptibility $\chi(T)$ is characteristic of antiferromagnetic and reproduces maximum in $\sigma(T)$ dependence.

3.2. Mössbauer spectroscopy

In Mössbauer spectra of all alloys, except for Fe73.5Al26.5 sample, the fast growth of fraction of non-magnetic component $P_0$ resulting in a decrease of the average $^{57}$Fe HFMF $\bar{H}$ at temperature from 12 to 120 K is observed (figures 2 and 3). Even for the alloy Fe73.5Al26.5 in [0-0.9]T/$T_c$ the dependence $\bar{H}(T)$
is different from analogous of ferromagnetic alloy Fe$_{70}$Al. In the Fe$_{70}$Al$_{30}$ samples the $\bar{H}$ increases at $T > 120$ K reaching maximum at 220-230 K regardless of the type of superstructure. The difference in minimal (at 110-130K) and maximal (at 220-230K) $\bar{H}$ values is $\approx 15$ kOe for the B2 and $\approx 25$ kOe for the D0$_3$ superlattice alloys (beyond the range of the error). Similar behaviour of $\bar{H}$ ($T$) for the alloy Fe$_{70}$Al$_{30}$ was observed also in [11].

The main feature of Mössbauer spectra taken under $H_{ext}$ is an appearance of the component with magnetic splitting highly exceeding an external field. Expanding of hyperfine magnetic splitting in spectra with $H_{ext}$ raise results in the essential increase of $\bar{H}$ (for the alloy Fe$_{65.9}$Al$_{34.1}$ at $T=80$K it is equal to 64 kOe under $H_{ext}$=16.2 kOe) [12]. The considerable difference in the temperature behaviour of $\bar{H}$ and $\sigma(T)$ vanishes at low values of $H_{ext}$ and these dependences get correlating. The temperature dynamics as well as in-field dynamics of Mössbauer spectra parameters testify the existence of relaxation phenomena in hyperfine interactions at $T>30-50$K (figures 2 and 3). But, the blocking temperature estimated from $\sigma(H/T)$ dependence is about 120-150K for Fe$_{65.9}$Al$_{34.1}$ and 210K for Fe$_{70}$Al$_{30}$ alloys [12]. What kind and size inhomogeneities could lead to collective spin fluctuations and to superparamagnetic signs we consider within the microscopic model of Fe local magnetic moment.

In [9] a simple model was proposed, in which the magnetic moment of an Fe atom surrounded up to 4 Al atoms is parallel, and of an Fe atom having 5 and more Al atoms in the nearest neighbourhood is antiparallel to the total magnetization. The model describes quantitatively the dependence of the Fe atom average magnetic moment on Al content and agrees with the data [6,7] on magnetic inhomogeneities on a nanometer scale. Inhomogeneity implies regions with magnetic moments directed along $m^+$ and opposite $m^-$ the total magnetization. Theoretically, the temperature dynamics of these $m^+$ and $m^-$ magnetic moments is different in consequence of Stoner-type excitations [13]. Suppose that thermal fluctuations of the $m^+$ magnetic moments, arising at rather low temperature (50-100 K), excite the fluctuations of the surrounding $m^+$ magnetic moments also. That results in collective spin fluctuations and abrupt decrease of $\bar{H}$ at $T\approx 100-120$K. Concurrent reduction and vanishing of the $m^+$ magnetic moments in absolute magnitude (due to Stoner-type excitations), that decrease faster than $m^-$, at 150<$T$<220 K brings to a gain of the total magnetization, to stabilization of surrounding $m^-$ magnetic moments and to the increase of $\bar{H}$.
Another reasoning equivalent to the above in relation to the obtained results based on two possible states of this magnetic system: with the local ferromagnetic and antiferromagnetic-type arrangement of neighbouring magnetic moments [8], is also possible. The theoretical studies [8,13] of metal-metalloid alloys have shown, that for high metalloid content the local magnetic moments opposite to magnetization change their direction with increasing the temperature. In this connection, the increase of $H$ at 220-230K can be explained by the augmentation of the number of magnetic moments $m^+$, with the magnitude being higher than predecessor moments $m^-$. So, the system goes into “high-spin” state. It leads to the gain in magnetization, enlargement of the ferromagnetic clusters size, reinforcement of the inter-cluster exchange, slowing down of the cluster magnetic moment fluctuations and growth of the magnetic hyperfine splitting in spectra. But, in this case, according to [8,13] one should expect the intensive maximum, localized in the narrow temperature range that is not observed in magnetometry.

By the results of the work [8], it is the state with spin-spiral wave, which corresponds to a minimal energy of the system. Then, the sharp collapse of hyperfine magnetic splitting from $T>$30-50K can be brought about by the destruction of spin-spiral wave state in favor of the state with Fe magnetic moments oriented parallel and antiparallel to magnetization. On further increasing temperature the effect of both, Stoner- type excitations or transition of the system in “high-spin” state, consists in the stabilization of the clusters fluctuations and disordered ferromagnetic spin structure formation. In this case, the coherence length of the spin spiral in the state with the spin-spiral ordering is the frame of the clusters in the state with the collinear spin ordering.

4. Conclusion
The results of temperature and in-field magnetometric and Mössbauer studies of the ordered Fe$_{100-x}$Al$_x$ ($25<x<35$ at.%) alloys give an indication of the collective spin fluctuations connected with the magnetic inhomogeneities on a nanometer scale. Temperature behaviour of the magnetization and the average $^{57}$Fe HFMF in 100-200K is assigned to several causes. One of explanations supposes “freezing” the relaxation phenomena in clusters consisting of spins oriented conversely the magnetization. To clarify which variant of the thermodynamic behaviour of the spin structure is most probable a quantitative analysis within itinerant magnetism thermodynamics is required.

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