

Investigations of short range ordering in Fe-(5-6)% (at.) Si single crystals with diffusive magnetic anisotropy

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The local atomic structure of the Fe-6 % (at.) Si alloy in samples subjected to various treatments (quenching, annealing in dc and ac magnetic fields) has been studied by X-ray diffraction and Moessbauer spectroscopy. In all the samples (even quenched from the disordering temperature of 850°C), the short-range ordering of the B2 type (CsCl structure) takes place at the average cluster size of 0.6–0.7 nm. No DO₃ ordering has been observed. The superfine field distributions calculated from NGR spectra have shown that the samples are ordered after all the treatments; moreover, a structure separation over the short-range ordering has been observed. The structure is formed by iron lattice where the short-range ordering regions are distributed consisting of single or doubled in the <100> direction (paired) bcc cells centered by silicon atoms. The number of Si atoms involved in the pair formation has been estimated.

Локальная атомная структура сплава Fe-6 ат.% Si в образцах, подвергнутых различным обработкам (закалке, термомагнитной обработке в постоянных и переменных магнитных полях), изучалась методами рентгеновской дифракции и мессбауэровской спектроскопии. Во всех образцах (даже после закалки от температуры разупорядочения 850°C) имеет место ближнее упорядочение по типу B2 (структура CsCl) со средними размерами кластеров 0,6–0,7 нм. DO₃-упорядочение не обнаружено. Распределения сверхтонких полей, рассчитанные из ЯГР-спектров, показали, что образцы после всех обработок упорядочены, кроме того, наблюдается расслоение в ближнем порядке. Структура сформирована решеткой железа, в которой распределены области ближнего упорядочения, состоящие из единичных или двояных (пары) в направлениях <100> ОЦК ячеек, центрированных атомами кремния. Сделана количественная оценка числа атомов кремния, участвующих в образовании пар.

Atomic structure and phase composition of soft ferromagnetic Fe-Si alloys have been investigated repeatedly and very thoroughly [1–3]. At Si content exceeding 7 % (at.), the DO₃ type ordering with antiphase domains was reported in the alloys. The domain size grows from 1–2 nm at 7.6 % (at.) Si to 20 nm at 20 % (at.) Si. At 5–6 % Si, only short-range ordering was observed [2], that was defined later as B2 one [4]. In the alloy with 8 % (at.) Si, we have discovered for the first time the co-exist-

ence of two phases: small locally ordered clusters of the B2 phase and up to 2 nm size domains of the DO₃ type ordering. In [4, 6], it is shown that in the Fe-(5–6) % (at.) Si alloy single crystals, an induced magnetic anisotropy formed upon annealing and cooling in dc field is accompanied by a uniaxial anisotropy of B2 clusters along the magnetic anisotropy direction, the average size of clusters reaching 0.9 nm.

This work presents the investigation results of single-crystalline samples Fe-6 % (at.) Si

Table 1. Samples and treatments

No.	Sample	Heat treatment		
		Type	Temperature	Magnetic field, A/m
1	G_q	quenching	from 850°C, 400°/s	internal, local
2	$G_{dc} l$	FA	450°C	10^6 , $H_{dc} \parallel [001]$
3	$G_{ac} l$	FA	450°C	$5 \cdot 10^4$, $H_{ac} \parallel [001]$
4	$G_{ac} t$	FA	450°C	$5 \cdot 10^4$, $H_{ac} \perp [001]$

with Goss orientation (only one easy direction [001] lies in the sample plane, the other two are directed at an angle of 45° to the plane) subjected to various treatments. During those treatments, the magnetic domain structure behaves in different manners. In a dc field, the sample is annealed substantially in the single-domain state. In contrast, in an ac (50 Hz) field, the domain walls move and become fragmented if the ac field is applied along [001] direction (from here on, longitudinally), and if the field is directed across [001] (from here on, transversally), the domain structure behavior is much more complex. Magnetic properties of samples after various treatments are different [7, 8]. This work is a continuation of X-ray investigations of atomic structure of single-crystalline Fe-6 % (at.) Si alloy. For additional information on the short-range atomic ordering, the nuclear gamma-resonance (NGR) spectroscopy is applied.

The X-ray diffraction is a powerful method sensitive to the peculiarities of both long-range and short-range structure ordering. While long-range ordering manifests itself in distinct diffraction peaks of a diffracted intensity, the correlations in distribution of neighboring atoms by type and/or their displacements from ideal positions in a distorted lattice results in considerable intensities of broad diffusion scattering peaks. It is just the measuring of the latter that provides the experimental data which, as being processed by modern computation methods, makes it possible to obtain information on short-range ordering in the atomic arrangement.

The method of NGR spectroscopy allows one to determine probabilities of local coordination and, comparing those with the statistic distribution of atoms in an alloy, judge the crystalline structure and the ordering processes.

The samples were cut out of a coarse-grained Goss electrotechnical steel Fe-6 % (at.) Si and shaped as thin single-crystalline disks. To refine all the samples, those were

annealed in vacuum at 1150–1300°C for 2–6 h, after which the carbon content was no more than 0.005 % (mass). Then, the samples were subjected to different treatments in air: quenching from 850°C at a rate of 400 grad/s, field-annealing (FA) from 450°C in a longitudinal dc field of 10^6 A/m, and in longitudinal and transversal ac fields of $5 \cdot 10^4$ A/m. The samples characterization is presented in Table 1.

For the X-ray diffraction examination, each sample was ground down to 100 μm and chemically etched to a thickness of 40–50 μm , which is optimum for Mo K_α radiation. The samples were examined at room temperature using a four-circle X-ray diffractometer with an X-ray tube and monochromator. An energy-dispersion detector registered intensity of scattered radiation. To eliminate completely the contribution to intensity from the main reflection formed by radiation with multiple harmonics ($\lambda/2$, $\lambda/3$, etc.), the anode voltage was lowered to 29 kV at which the $\lambda/2$ bremsstrahlung is not excited at all. We used the transmission geometry for the incident X-ray beam to pass through a thin sample. This weakens the intensity but allows us to perform measurements in a wide range of reciprocal space and, in particular, to measure any reflections with a scattering vector lying in the sample plane. For the same samples, the NGR spectra were measured. In the course of spectra computation, a modelless fitting with 7–8 sextets with the line width close to the one in α -Fe spectrum was performed. Such procedure allowed us to gain complete spectra description under condition of minimum possible mean-square deviation from the experiment.

For all samples, diffuse scattering along [001] was measured upon scanning over the scattering angle θ –2 θ , which exhibited broad diffuse maxima corresponding to (001) and (003) reflections forbidden in the bcc lattice, Fig. 1. These maxima are forbidden in pure iron and appear in the presence

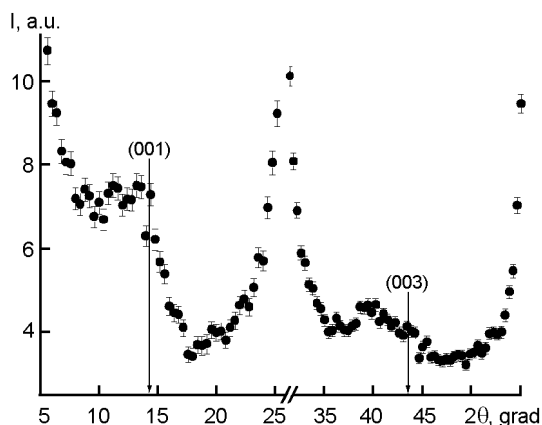


Fig. 1. Intensity profiles upon θ - 2θ scanning of diffuse scattering.

of substitution impurities, for example, Si atoms in iron. The presence of diffuse maxima in the sites with an odd sum of h , k , l indices and their absence in the sites with half-integer indices evidence the B2 ordering and absence of DO_3 ordering. It is of interest that the observed B2 superstructure reflections are shifted from the ideal positions towards smaller dispersion angles [5]. This fact testifies to local lattice distortions around Si atoms. From the half-width of the diffuse reflections, we can estimate the average size of regions contributing to this reflections [9]. The results of such procedure are given in Table 2. After quenching from the disordering temperature (850°C) estimated in [2], the ordered regions (clusters) have the average size along [001] of about 0.6 nm, for other samples this size was somewhat larger than 0.7 nm, Table 2. This means that the cluster includes on average two elementary cells with one common face and one Si atom in the center of each cell.

Fig. 2 represents distributions of contributions to the spectral-line intensities from different Fe atom coordinations, determined from the NGR spectra fitting for the samples after all the above treatments. It is seen that treatments influence the distribu-

Table 2. Average size of B2 clusters along [001] direction

No.	Sample	Size, nm
1	G_q	0.59 ± 0.06
2	$G_{dc} l$	0.72 ± 0.02
3	$G_{ac} l$	0.74 ± 0.02
4	$G_{ac} t$	0.72 ± 0.02

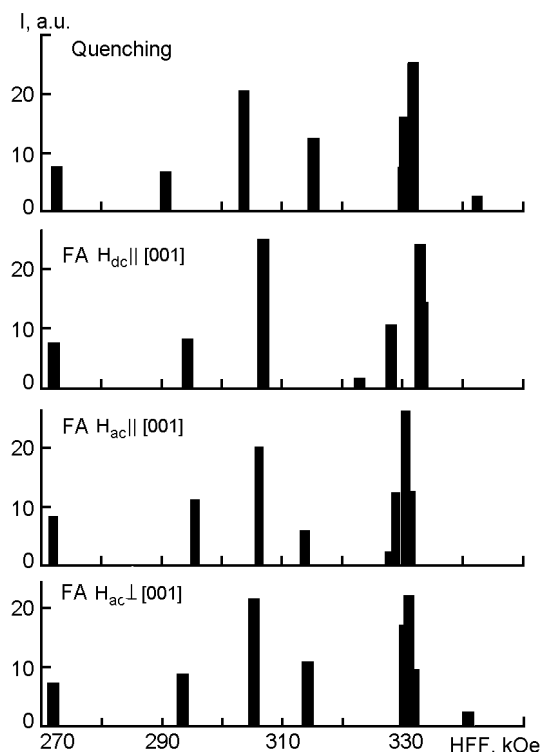


Fig. 2. Histograms of contributions to spectral intensities from configurations with different hyperfine fields.

tion type. The lines with the field of 317 to 342 kOe belong to the 8:0 configuration (8 Fe atoms and 0 Si atoms in the first coordination shell, respectively). According to [10], additional lines belong to atomic configurations where Si atom is absent in the 2th through 5th shells as well. Coordination with one Si atom in the first shell is divided into two lines – 7:(10) and 7:(11), where (10) and (11) mean the occupancy of the nearest, namely, first and second coordination shells by Si atoms. This coordination corresponds to one Si atom in the first sphere and one Si atom in the first and in the second spheres, with corresponding fields 302 and 291 kOe, respectively. The attempt to subdivide this line into its constituents similarly to 8:0 coordination were unsuccessful. This suggests that the additional lines for the 8:0 coordination are connected with the Fe regions of a size at least 2 to 4 lattice parameters. For the single-phase alloy, the 8:0 and 7:1 configurations must be described by the same number of lines. The difference in description points to the existence of different phases. From the intensity ratio of 7:1 and 6:2 lines, assuming that it is no ordering in Fe-Si phase, the Si content was determined to be

Table 3. Intensity ratio of superfine fields as calculated from NGR spectra

No	Sample	I_1/I_0 , ± 4 %	I_2/I_0 , ± 4 %	I_{11}/I_{10}	I_2/I_1
1	G_q	0.43	0.12	0.33	0.28
2	$G_{ac l}$	0.58	0.14	0.33	0.23
3	$G_{dc l}$	0.52	0.14	0.56	0.27
4	$G_{dc t}$	0.49	0.12	0.41	0.24

about 7 % (at.). At the same time (see Table 3), the ratios of line intensities for 8:0 (I_0), 7:1 (I_1) and 6:2 (I_2) configurations, where I_0 is the total intensity of lines without Si atoms; I_1 , the sum of intensities with one Si atom; and I_2 , the same with two Si atoms in the first coordination shell around Fe, change in the course of treatment. This seems to be connected with changes in the volume of regions depleted of Si. An increase of the I_{11}/I_{10} ratio for Sample 3 points to increase in the occupancy of (11) type sites, which may be interpreted as the presence of ordering.

Annealing in longitudinal and transversal ac fields results in different values of intensity ratios (Table 3). For both these treatments, I_1/I_0 , I_2/I_0 , and I_2/I_1 ratios are essentially equal to each other, but I_{11}/I_{10} differs noticeably. If the annealing is performed in a longitudinal ac field, the probability of Si atom appearance in the second shell in the presence of one Si atom in the first one increases remarkably. This is an evidence for the ordering process intensification after the treatment in longitudinal ac field in comparison with that in the transversal field.

In coordination 7:1 with configuration (01), one Si atom occupies one of the cube vertices around Fe atom. In this case, the configuration (11) may be formed by the second Si atom which is either the nearest to the first Si atom neighbor in the $\langle 111 \rangle$ direction, or Si atom in most possible distant position in the $\langle 113 \rangle$ one. As the first situation is impossible (Si-Si atom pair in Fe-Si alloys is never formed by the nearest neighbors in the $\langle 111 \rangle$ direction), the second situation is realized most likely, which means that Si atoms are spaced at a distance of the fourth neighbors. If the induced magnetic anisotropy is really connected with the distribution anisotropy of Si atoms along main $\langle 100 \rangle$ directions, then the configuration (11) cannot contribute to the induced magnetic anisotropy. As to the

samples quenched and dc field-annealed, those both are in the IMA state (in the course of quenching, the field-annealing in a local field in each magnetic domain took place). For these samples, the I_{11}/I_{10} ratio is essentially lower compared to that for the ac field-annealed samples. After the ac field-annealing, the relative number of (11) configurations of Si-Si atomic pairs oriented in the $\langle 113 \rangle$ direction increases. Both these configurations cannot take part in the magnetic anisotropy inducing. So, the ordering process in the course of the ac field-annealing does not favor magnetic anisotropy inducing.

In 6:2 coordinations (two Si atoms in the first shell), Si-Si pairs can be oriented along the cube ribs ($\langle 100 \rangle$ direction), face diagonals ($\langle 110 \rangle$) or cube diagonals ($\langle 111 \rangle$). According to the diffraction data, the first variant is statistically most probable, since the extension of B2 clusters along [001] direction is somewhat more than the sum length of two cells (the lattice parameter is 0.2862 nm). In this case, the nearest surrounding of Si-Si pair contains 12 Fe atoms, only 4 of those having configuration of the 6:2 nearest surrounding, while the other, 7:1. The relative intensities must relate as $I_2:I_1 = 1:2$. In fact, from the NGR spectroscopy data, the ratio $I_2:I_1$ ranging from 0.24 to 0.28 is obtained. So, the relative amount of Si atoms in pairs along [001] is of 48 to 56 %, or about half.

Thus, the atomic structure of the Fe-6 % (at.) Si alloy can be represented as a bcc lattice of the FeSi alloy depleted of silicon, where the B2 type ordered clusters are incorporated. Depending on the sample magnetic state and treatment routine, the following specific features can be picked out: the state with the maximum magnetic anisotropy extent, which is formed under annealing in a dc magnetic field and is characterized by the maximum degree of structure separation; field annealing in an ac magnetic field results in an increased ordering degree which is not related to the formation of magnetic anisotropy induced upon field annealing in the dc magnetic field, with the maximum degree of ordering gained upon longitudinal application of the ac field; the relative content of Si atoms that form pairs of directional B2 ordering along $\langle 100 \rangle$ axes amounts 48 to 56 %, with the maximum fraction corresponding to the samples either quenched or annealed in the ac field applied along the easy axis, and the minimum one

in samples subjected to field annealing in the longitudinal dc and transverse ac fields.

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Дослідження ближнього порядку у монокристалах Fe–(5–6) % (ат.) Si з дифузійною магнітною анізотропією

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Локальну атомну структуру сплаву Fe–6 % (ат.) Si у зразках, підданих різним обробкам (загартування, термомагнітна обробка у постійних та змінних магнітних полях) досліджено методами рентгенівської дифракції та мессбауеровської спектроскопії. У всіх зразках (навіть після гартування від температури розупорядкування 850°C) має місце близьке впорядкування за типом B2 (структура CsCl) з середніми розмірами кластерів 0,6–0,7 нм. DO₃-впорядкування не виявлено. Розподіли надтонких полів, розраховані з ЯГР-спектрів, посвідчили, що зразки після усіх обробок є впорядкованими, крім того, спостерігається розшарування у ближньому порядку. Структура сформована ґраткою заліза, в якій розподілені області ближнього впорядкування, які складаються з поодиноких або здвоєних (пари) у напрямках <100> ОЦК комірок, центрованих атомами кремнію. Зроблено кількісну оцінку кількості атомів кремнію, що беруть участь в утворенні пар.