MAGNETIC STRUCTURE OF FACE-CENTERED CUBIC NICKEL-IRON ALLOYS

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The presence of a spatially inhomogeneous magnetic structure in face-centered cubic nickel alloys is demonstrated by investigating magnetic elastic incoherent neutron scattering. The experimental results are explained theoretically.

INTEREST in the magnetic structure of fcc iron-nickel alloys arose in connection with the problem of invar properties. The fact that this structure is not trivial is indicated by the unusual behavior of the average magnetic moment per atom of the alloy, $\bar{\mu}(c)$. Kondorskiĭ and Fedotov[1], and later Grangle and Hallam[2] have shown experimentally that $\bar{\mu}(c)$ increases linearly up to a concentration of 50 at.% iron in nickel, and then decreases rapidly in the concentration interval 50–75 at.% iron (Fig. 1).

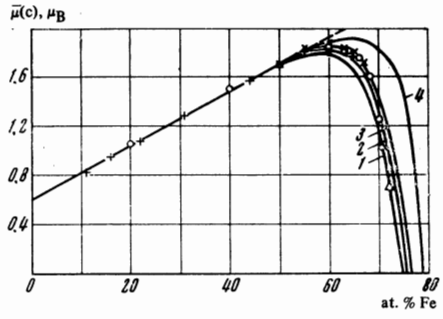

FIG. 1. Dependence of the average magnetic moment per atom on the composition in fcc iron-nickel alloys: +–data of Kondorskiĭ and Fedotov [1], O–data of Grangle and Hallam [2], X–data of present research, Δ–data of Bando [16]. Solid curves–theoretical plots: 1–for perfectly disordered alloys, 2–for alloys with correlation $\epsilon_{22} = -0.01$, 3–for alloys with correlation $\epsilon_{22} = -0.02$, 4–for alloys with the order of Fe_3Ni ($\eta* = 1$ and $\epsilon_{22} = 0$).

It can be assumed that such a behavior of $\bar{\mu}(c)$ is due to the change of the atomic magnetic moments of the iron and of the nickel as a result of a redistribution of the electrons in the unfilled 3d shell. However, investigations of the x-ray spectra[3] have shown that the structure of the energy bands is deformed very little as a function of the composition and it is difficult to expect so strong a redistribution of the electrons in a small concentration interval (10–15%).

In our opinion, a more fruitful approach to explanation of the magnetic structure of fcc iron-nickel alloys was first indicated by Kondorskiĭ and Sedov[4,5], who proposed on the basis of an experimental study of the temperature dependence of the magnetic susceptibility of austenitic steel that in fcc iron alloys there exists an antiferromagnetic interaction between the iron atoms (the so-called "latent antiferromagnetism"). This hypothesis was subsequently confirmed experimentally in experiments on small-angle inelastic scattering of neutrons[6].

Developing this idea further, Sidorov and Doroshenko[7,8] proposed that by virtue of the competition between the ferro- and antiferromagnetic interactions in iron-nickel alloys, a noncollinear magnetic structure is produced, in which the atomic magnetic moments, regarded as classical vectors, are rotated through different angles relative to the direction of the spontaneous magnetization. Consequently, the magnetization of the alloy is determined by the sum of the projections of the magnetic moments of the alloy atoms on this direction. The decrease in the magnetization of the alloy with increasing content of the "antiferromagnetic" component (in our case, iron) is due to the decrease of these projections as a result of the increasing disorientation of the atomic magnetic moments. The statistical fluctuations of the nearest surroundings of the atoms determine in this case unequal "turnings" of the atomic magnetic moments at different sites. As a result, the magnetic structure turns out to be inhomogeneous. On the basis of this model, the authors of[7,8] succeeded in describing quantitatively the experimentally observed concentration dependence of $\bar{\mu}$ of iron-nickel alloys.

In[9], Izyumov and Medvedev have shown that a quantum-mechanical consideration of the problem confirms the validity of this classical model. It follows also from this paper that a quantum-mechanical allowance for the antiferromagnetic interaction of the pairs of atoms in the ferromagnetic matrix leads to a local cancellation of the z-projections of the spins of the pair atoms and of the atoms located near such a pair. Consequently, the magnetic structure of the alloy remains collinear, but not uniform from site to site of the crystal lattice. An important factor here is that local cancellation of the z-projections does not take place unless the antiferromagnetic interaction exceeds a definite value.

It is known from general diffraction theory that if the radiation is scattered by inhomogeneous media, then incoherent small-angle scattering is observed in the diffraction pattern, and the magnitude of the effect does not depend on the sign of the inhomogeneities. One could therefore hope that magnetic diffuse small-angle scattering of neutrons would make it possible to observe experimentally the effect due to the inhomogeneity of the magnetic structure of iron-nickel alloys, which, as noted above, follows from an analysis of the classical and quantum-mechanical models of these alloys. This has been the aim of the present investigation.

1. THEORY

The theory of magnetic diffuse scattering of neutrons by alloys having spatial inhomogeneity was considered by Marshall[10]. The differential cross section of magnetic diffuse scattering is given by

$$\left(\frac{d\sigma}{d\Omega}\right)_d = \frac{N}{4}(r_0\gamma)^2[1-(\mathbf{em})^2]T(\mathbf{K}), \quad (1)$$

where

$$T(\mathbf{K}) = \frac{1}{N}\sum_{n,m}\exp\{i\mathbf{K}(\mathbf{R}_n-\mathbf{R}_m)\}(\overline{f_n f_m}-\bar{f}^2). \quad (2)$$

Here r_0 is the electromagnetic radius of the electron, γ the magnetic moment of the neutron, **e** the unit vector of scattering, **m** the unit vector of the magnetic moment, f_n and f_m the magnetic scattering amplitudes of atoms of species n and m, $\bar{f}$ the average scattering amplitude, N the number of atoms in the crystal, and $K = 4\pi\lambda^{-1}\sin\theta$.

Marshall has shown that in the nearest-neighbor approximation, with allowance for the atomic correlation, the differential forward-scattering cross section is

$$T(0) = c(1-c)\left(\frac{\partial\bar{\mu}}{\partial c}\right)^2 + \frac{2c^2(1-c)^2}{Z}\left(\frac{\partial\bar{\mu}}{\partial\epsilon_{22}}\right)^2, \quad (3)$$

where c is the concentration of atoms of species 2 (in our case iron), Z the number of nearest neighbors, and $\epsilon_{22} = (p_{22}c - c^2)$ the correlation parameter, equal to the difference between the physical probability ($p_{22}c$) of replacing a pair of sites by atoms of species 2 in the given solution, and the probability of its replacement for a random distribution of the atoms over the lattice sites, which is equal to c^2; p_{22} is the probability that the lattice site closest to an atom 2 is occupied by an atom 2; ϵ_{22} can be either positive or negative if short-range order is present in the alloy.

To be able to compare the experimental data with the theoretical ones, it is necessary to know the analytic expression for $\bar{\mu}$ as a function of the concentration and of the correlation. We derive this expression by using the quantum-mechanical representation of the magnetic structure of alloys with mixed exchange interaction. We start from the fact that cancellation of the z-projection of the atomic spin and of the corresponding magnetic moment occurs only if atoms of species 1 (Ni) and 2 (Fe) are surrounded by a sufficient number of atoms of species 2. Accordingly, we assume that cancellation of the z-projection of the magnetic moment of an atom of species 2 will take place if among the nearest neighbors surrounding it there are at least X atoms of species 2. This means that the statistical fluctuations in which the number of atoms 2 that are closest to an atom 2 is equal to zero, 1, 2, ..., X − 1, should not be taken into account in the calculation of the cancellation of the z-projection of the magnetic moment of the atoms 2. We write down the average z-projection of the atomic magnetic moment of an atom of species 2 ($\mu_z^{(2)}$) as a quantity that depends on a certain effective probability of substitution of the site closest to the atom 2 by an atom of the same species (p'_{22}). In the nearest-neighbor approximation we have

$$\bar{\mu}_z^{(2)} = \mu_2 - \mu_2 g_2 p_{22}'(c), \quad (4)$$

where μ_2 is the atomic magnetic moment of the atom of species 2, and g_2 is a certain dimensionless coefficient. The quantity $\mu_2 g_2 p'_{22}(c)$ determines the decrease of μ_2 introduced on the average by all the surrounding nearest-neighbor atoms.

It is obvious that $p'_{22}(c) = p_{22}(c) - p''_{22}(c)$, where $p''_{22}(c)$ is the effective probability of replacing the site closest to the atom 2 by an atom of the same species, such that no cancellation of the z-projection of the magnetic moment of atom 2 takes place. The quantities $p_{22}(c)$ and $p''_{22}(c)$ can be calculated as mean values of the probability $p_Z(n, p_{22})$ that atoms 2 occupy n sites out of the Z nearest sites surrounding each atom 2:

$$p_{22}(c) = \frac{1}{Z}\sum_0^Z P_z(n,p_{22})n, \qquad p''_{22}(c) = \frac{1}{Z}\sum_0^{X-1} P_z(n,p_{22})n, \quad (5)$$

where X = 1, 2, ..., Z and the summation is terminated on the basis of the considerations advanced above. Here $P_Z(n, p_{22})$ satisfies the binomial distribution[11]

$$P_z(n,p_{22}) = \frac{Z!}{n!(Z-n)!}p_{22}^n p_{21}^{Z-n}. \quad (6)$$

The expression for the z-projection of the magnetic moment of an atom of species 1 ($\bar{\mu}_Z^{(1)}$) can be written in the form

$$\bar{\mu}_z^{(1)} = \mu_1 - \mu_1 g_1 p_{12}'(c)\,p_{22}'(c), \quad (7)$$

where μ_1 is the magnetic moment of the atom of species 1, g_1 is a constant coefficient, and $p'_{12}(c)$ is the effective probability of replacement of a site closest to the atom of species 1 by an atom of species 2. Since there is positive exchange interaction between atoms 1 and 2, the cancellation of μ will occur only when the atoms of species 2 situated in the nearest surrounding of atom 1 are themselves surrounded by a sufficient number of atoms of species 2. The probability of such a complicated event should be proportional to the product of the probabilities $p'_{12}(c)$ and $p'_{22}(c)$. Bearing in mind that $p'_{12}(c) = p'_{21}(c)c/(1-c)$ (which follows from the natural requirement $p'_{22}(c) + p'_{21}(c) = 1$), we write

$$\bar{\mu}_z^{(1)} = \mu_1 - \mu_1 g_1 \frac{c}{1-c}p_{21}'(c)\,p_{22}'(c). \quad (8)$$

We now find the average magnetic moment of the alloy:

$$\bar{\mu}(c) = \bar{\mu}_z^{(1)}(1-c) + \bar{\mu}_z^{(2)}c$$
$$= \mu_1(1-c)\left\{1 - g_1\frac{c}{1-c}p_{21}'(c)p_{22}'(c)\right\} + \mu_2 c\{1 - g_2 p_{22}'(c)\}. \quad (9)$$

Formula (9) goes over into the formula given for $\bar{\mu}(c)$ in[11] and derived from other physical premises, provided the following notation is used:

$$g_1 = Z^2\lambda_1\lambda_2, \qquad g = Z\lambda_2, \qquad \delta(p_{22}) = \frac{1}{Z}\sum_0^{X-1} P_z(n,p_{22})n. \quad (10)$$

Consequently

$$\bar{\mu}(c) = \mu_1(1-c)\left\{1 - Z^2\lambda_1\lambda_2\frac{c}{1-c}[p_{22}+\delta(p_{22})]\right.$$
$$\left.\times[1-p_{22}-\delta(p_{22})]\right\} + \mu_2 c\{1 - Z\lambda_2[p_{22}+\delta(p_{22})]\}. \quad (11)$$

For a perfectly disordered alloy $p_{22} = c$, and when correlation is taken into account $p_{22} = c + \epsilon_{22}/c$. Equation (11) contains three unknown parameters X, λ_1, and λ_2. We use for their determination the condition $\bar{\mu}(c) = 0$ at a certain concentration c_0, which is taken from experiment. Since the atomic magnetic moments are essentially positive quantities at any concentration c, the vanishing of $\bar{\mu}(c)$ is possible only if the coefficients of μ_1 and μ_2 are individually equal to zero:

$$1 - Z\lambda_2 p_{22}'(c_0) = 0, \quad 1 - Z^2\lambda_1\lambda_2 p_{22}'(c_0)\, p_{12}'(c_0) = 0. \qquad (12)$$

The third independent equation is also obtained from a consideration of the state of the system at the point c_0. Indeed, complete cancellation of the magnetic moments of species 1 at $c = c_0$ should occur when for each atom 1 (their number in the alloy is $N(1 - c)$) there is a definite number k of effective 2–2 pair interactions, the number of which is $Nc_0Zp_{22}'(c)$. Consequently, the following relation holds at the point c_0:

$$Nc_0 Z p_{22}'(c_0)/N(1 - c_0) = k.$$

Using this expression and (12), we get

$$\lambda_2 = kc_0/(1 - c_0). \qquad (13)$$

It turned out that in most cases encountered to date[7,8] agreement between the experimental and theoretical curves was observed at k = 1. For disordered iron-nickel alloys, X = 12 and $c_0 = 0.744$[8]. Consequently, expression (11) simplifies, since $\delta(p_{22}) = -p_{22}(1 - p_{22}^{11})$ and

$$\bar{\mu}(c) = \{1 - c - Z^2\lambda_1\lambda_2 c(c + \varepsilon_{22}/c)[1 - (c + \varepsilon_{22}/c)^{11}]\}\mu_1 + \{c - Zc(c + \varepsilon_{22}/c)^{12}\lambda_2\}\mu_2. \qquad (14)$$

Thus, the analytic expression (14) for $\bar{\mu}$ contains a dependence on the concentration c and on the correlation ϵ_{22}.

2. DESCRIPTION OF EXPERIMENT

For an experimental investigation of $\bar{\mu}(c)$ and $d\sigma/d\Omega = f(c)$, we chose eight alloys containing 25, 40, 50, 55, 60, 63, 65, and 67 at.% iron in nickel[1]. The alloys were smelted in vacuum using pure components. Deviations from the indicated compositions amounted to no more than 0.2 at.%. The carbon content was less than 0.01%. After prolonged homogenization (~ 100 hours) at 1100°C, the alloys were quenched in water. The purpose of such treatment was to obtain the most disordered state. The saturation magnetization was measured with a ballistic setup and with a vibromagnetometer in the range from room temperature to the temperature of liquid nitrogen and in fields up to 12 kOe. In the former case the samples were in the form of a cylinder 50 mm long and 5 mm in diameter, and in the latter in the form of disks 3.5 mm in diameter and 1 mm thick.

The diffuse magnetic scattering of neutrons was measured at liquid-nitrogen temperatures using a diffractometer mounted in the horizontal channel of the IVV-2 reactor. We investigated the scattering of neutrons passing through samples in the form of plates measuring 20 × 20 mm and 2 mm thick. The neutrons were made monochromatic by reflection from a bent single crystal of iron silicide[12]. The employed wavelength was 1.13 Å. The collimator in front of the sample was 5 mm wide and 12 mm high, making it possible to fix a minimum scattering angle $2\theta = 1°30'$. The magnetic scattering of the neutrons was separated with the aid of a magnetic field of 6 kOe intensity[2]. The corrections for multiple scattering were introduced by Vineyard's method[13,14].

3. EXPERIMENTAL RESULTS AND DISCUSSION

The lying crosses in Fig. 1 represent our experimental values of $\bar{\mu}(c)$, obtained for Fe-Ni alloys by extrapolating the temperature dependence of the saturation magnetization to 0°K in accordance with the $T^{3/2}$ law, although this law does not hold very accurately for these alloys[15]. The same figure shows the values of $\bar{\mu}(c)$ from[1,2,16]. Good agreement between our experimental data and the published ones is observed. The solid curve 1 shows the theoretical $\bar{\mu}(c)$ plot for perfectly disordered alloys, for which $p_{22} = c$, X = 12 and $c_0 = 0.744$ ($\lambda_1 = 0.0299$, $\lambda_2 = 2.9$). We see that in the region of compositions from 50 to 75 at.% Fe, the experimental values do not fit the theoretical plot. (This induced us to investigate this region in greater detail.)

Such a discrepancy between theory and experiment can be explained by recognizing that it is practically impossible to obtain iron-nickel alloys in a perfectly disordered state even at very high quenching rates. As shown experimentally, a short-range order is always present in them ($\epsilon_{22} < 0$)[3]. Allowance for the correlation of these alloys has resulted in good agreement between the theoretical $\bar{\mu}(c)$ plot and the experimental values.

Curve 2 of Fig. 1 was plotted for $\bar{\mu}(c)$ under the assumption that $\epsilon_{22} = -0.01$. In this case $c_0 = 0.754$, with $\lambda_1 = 0.0285$ and $\lambda_2 = 3.065$. Curve 3 was plotted under the assumption that $\epsilon_{22} = -0.02$ and $c_0 = 0.764$, with $\lambda_1 = 0.0265$ and $\lambda_2 = 3.228$. We see that at compositions up to 70 at.% Fe the experimental points fit well the theoretical curve with $\epsilon_{22} = -0.02$. For alloys with larger iron concentration, the data fit well a theoretical curve with smaller or zero correlation. The decrease of the correlation parameter with increasing iron concentration is confirmed by experiments on diffuse neutron scattering.

Curve 4 of Fig. 1 shows the theoretical dependence of the average magnetic moment for alloys having the same order as Fe_3Ni with an ordering degree $\eta_* = 1$ and with zero correlation. We assumed here that in formula (11) $p_{22} = c - \gamma^2\eta_*^2/c$, where $\gamma = 1/3$ for $c \leq 3/4$ and $\gamma = 1 - c$ for $c \geq 3/4$.

Our attempts to obtain an ordered state of the Fe_3Ni type in alloys of near-stoichiometric composition ended in failure. Apparently no such superstructure exists in the Fe-Ni system.

The experimental angular dependences of magnetic diffuse scattering of neutrons are shown in Fig. 2. We see that for alloys in the composition region where the law of mixing of the magnetic moments is satisfied, the diffuse-scattering curves have a clearly pronounced intensity dip near small K. At the same time, for alloys in the region of the sharp variation of the average magnetic moment, a significant increase of the small-angle scattering intensity is observed, and this effect increa-

[1] Unfortunately, owing to the presence of a martensitic transformation above the nitrogen temperature, we were unable to investigate alloys with iron concentrations exceeding 68 at.%.

[2] This field was possibly insufficient for complete saturation of samples in the form of a plate. Our results are therefore somewhat too low. This, however, does not change the qualitative picture of the effect at all.

[3] With the aid of nuclear diffuse scattering of neutrons, using the Ni^{62} isotope, we determined experimentally the sign and magnitude of the correlation in quenched fcc Fe-Ni alloys in the region of concentrations with the anomalous behavior of $\bar{\mu}(c)$. The correlation turned out to equal ~0.01–0.02, and its negative sign indicated the presence of short-range order in these alloys. Details will be reported separately.

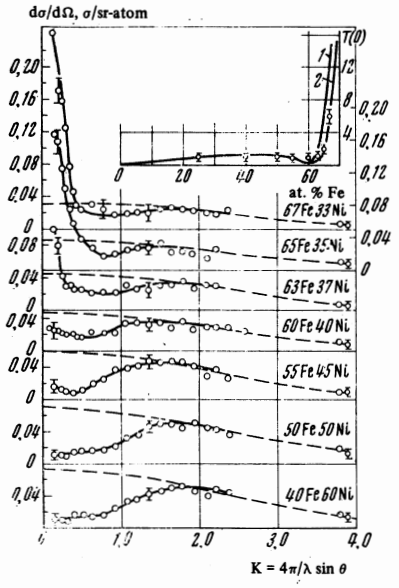

FIG. 2. Experimental angular dependences of the cross section for the magnetic diffuse scattering of neutrons for different alloys. The insert shows the concentration dependence of the forward scattering cross section at 0°K. Solid curves—theoretical plots: $1-\epsilon_{22} = 0$, $2-\epsilon_{22} = -0.02$. The light circles show the experimental values.

ses with increasing content of the iron in the alloy. An examination of Fig. 2 shows also the transformation that occurs in the angular dependence of diffuse magnetic scattering as a function of the iron concentration in the alloy. Unfortunately, owing to experimental difficulties, it is impossible to come sufficiently close to a zero scattering angle, and the procedure of extrapolation to the zero angle remains somewhat arbitrary. Nonetheless, in many cases it is possible to determine T(0) with sufficient accuracy.

The light circles of Fig. 2 show the experimental results for the concentration dependence T(0) obtained by extrapolating the angular dependence of the magnetic neutron scattering cross section to K = 0. In the upper part of the same figure, the solid curves 1 and 2 represent the theoretically-calculated values of T(0) in accordance with formula (3), assuming $\epsilon_{22} = 0$ (curve 1) and $\epsilon_{22} = -0.02$ (curve 2). The experimental values of T(0) fit well the curve 2, leading to the conclusion that these alloys have negative correlation.

The experimental values of T(0) for the alloys with 25, 40, 50, and 55% were obtained by extrapolating the form-factor dependence of the magnetic scattering cross section from the region of large K. Such a procedure was used because the $d\sigma/d\Omega$ curves of these alloys, as functions of K, are distorted by the short-range magnetic order, which duplicates the short-range nuclear order, and also because the measurements of $(d\sigma/d\Omega)_d$ were not made at a temperature close to 0°K, but at 100°K. The latter circumstance is very important and will be considered by us in our next paper.

It is known that the region of large K on the angular dependence $(d\sigma/d\Omega)_d$ is least distorted by extraneous phenomena, and the experimental values agree well with the theoretical plot of the square of the form factor. Using this fact, we can obtain the values of $(d\sigma/d\Omega)^0$ extrapolated to K = 0. These curves are shown dashed

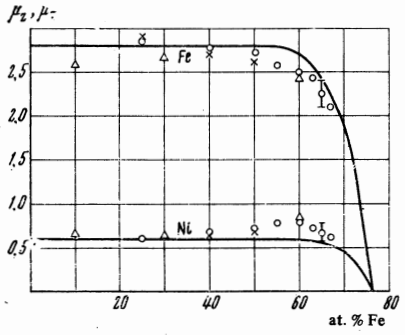

FIG. 3. Concentration dependences of the z-projections of the atomic moments of iron and nickel. Solid curves—theoretical. Points—experimental: X—[17], Δ—[18], O—present work.

in Fig. 2. By considering simultaneously the data on the magnetic moment $\bar{\mu}(c)$ and $(d\sigma/d\Omega)^0$, we can determine the magnetic moments of the Fe and Ni atoms. Putting $\epsilon_{22} = 0$, and also assuming the mixing law to be satisfied for the cancelled magnetic moments in the entire concentration region, we have

$$\left(\frac{d\sigma}{d\Omega}\right)^0 = 0.073c(1-c)[\bar{\mu}_z^{(2)} - \bar{\mu}_z^{(1)}]^2.$$

Using this expression and relation (9), we can obtain the experimental values of $\bar{\mu}_z^{(1)}$ and $\bar{\mu}_z^{(2)}$, which depend on the concentration. These values are shown by the light circles in Fig. 3. In the concentration region from 0 to 50 at.% Fe, the values of $\bar{\mu}_z^{(1)}$ and $\bar{\mu}_z^{(2)}$ agree well with the magnetic moments of the iron atoms ($\mu_2 = 2.8\ \mu_B$) and nickel atoms ($\mu_1 = 0.6\ \mu_B$) determined earlier in[15], whereas in the composition region from 50 to 75 at.% they disagree significantly. Consequently, the magnetic neutron scattering occurs on the average z-projection of the atomic spin. Figure 3 shows a comparison of the experimental and theoretical plots of $\bar{\mu}_z^{(2)}$ and $\bar{\mu}_z^{(1)}$, calculated from expressions (9) and (11). Full agreement between them could hardly be expected, owing to the large experimental errors, but the general character of the variation of $\bar{\mu}_z^{(2)}$ and $\bar{\mu}_z^{(1)}$ with the concentration can be traced experimentally quite correctly.

CONCLUSION

The magnetic small-angle scattering of neutrons observed in the present investigation points to the existence of an inhomogeneous magnetic structure in fcc iron-nickel alloys. This structure can be regarded as collinear if account is taken of only the z projections of the magnetic moment of the atoms on the direction of the spontaneous magnetization, and noncollinear if the total magnetic moment is considered. Since the cancellation of the atomic magnetic moment depends on the nearest surroundings, owing to the presence in the fcc iron-nickel alloys of a mixed exchange interaction ($I_{Fe-Fe} < 0$, $I_{Fe-Ni} > 0$, $I_{Ni-Ni} > 0$), it follows that by virtue of the statistical distribution of the atoms the quantum-mechanical z-projection of the atomic spin assumes different values from site to site. The degree of spatial magnetic inhomogeneity increases when the iron concentration approaches the point $c_0 = 0.75$.

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