

THE MÖSSBAUER EFFECT IN FACE-CENTERED CUBIC LATTICES OF IRON-NICKEL ALLOYS

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The effective field strengths on Fe^{57} nuclei in the fcc lattices of iron-nickel alloys are measured as functions of concentration and temperature by the nuclear γ resonance technique. The assumption underlying the explanation of $H_{\text{eff}}(c, T)$ in the alloys is that the effective field on the nucleus and the effective molecular field are proportional to each other ($H_{\text{eff}} \sim H_{\text{mol}}$), the latter field being created by atoms of the immediate neighborhood on the iron atom. The variation of $H_{\text{eff}}(c)$ in the iron-nickel alloys is related to the coexistence of positive and negative exchange interactions between atom pairs ($I_{\text{Fe-Fe}} < 0$, $I_{\text{Fe-Ni}} > 0$, $I_{\text{Ni-Ni}} > 0$). The dependence of the effective field on the nucleus on temperature and degree of ordering in the alloy can also be explained on basis of the concepts employed here.

FOLLOWING the discovery of γ resonance on Fe^{57} nuclei^[1], it became possible to use the Mössbauer effect to study the magnetism of metals and alloys. The very first experiments^[2] on pure iron have shown that the effective field at the Fe^{57} nucleus has the same temperature dependence as the saturation magnetization. This led to the important conclusion that the frequency of the Larmor precession of the nuclear spin (ω_L) is much lower than the frequency of the flipping of the electron spin (ω_s), and the Zeeman splitting of the nuclear levels is consequently determined by the mean value of the atomic spin. It was natural to assume that the effective magnetic field at the Fe^{57} nucleus (H_{eff}) is proportional to the average magnetic moment per atom ($\bar{\mu}$) not only for the pure metal but also for its alloy with other elements. An experimental verification of this was first obtained by Johnson et al.^[3] for two-component systems of iron with Al, Si, Co, Ni, V, and Cr. It turned out that the concentration dependences of H_{eff} and $\bar{\mu}$ are very similar but there is no proportionality between them. An appreciable discrepancy was observed between the concentration gradients of these quantities. In addition, the effective field at the Fe^{57} nucleus turned out to be very sensitive to the nearest surrounding of the atoms. All this has suggested that investigations of the effective magnetic fields at the Fe^{57} nuclei lead new information (not duplicating the results of magnetic measurements) concerning the nature of magnetism of metals and alloys.

In the present study, we used the Mössbauer effect to investigate the magnetic state of atoms in iron-nickel alloys with fcc lattices, for the purpose of obtaining additional information concerning the magnetic structure of invar alloys, which was also investigated by the magnetic neutron diffraction method^[4]. Earlier investigations^[3] of this system were made mainly at room temperature and have shown first an increase of H_{eff} with increasing iron concentration in the nickel up to 55 at.%, followed by a steep decrease to zero at 70 at.% iron. Such a concentration dependence of the effective field at the Fe^{57} nucleus is quite similar to the $\bar{\mu}(c)$ de-

pendence, but the comparison was insufficient, since values of H_{eff} extrapolated to 0°K were not available for all the alloys. In addition, Japanese papers published subsequently^[5,6] reported quite contradictory results both with respect to the magnitudes of the effective fields and with respect to the forms of the spectra of alloys in the composition range 65–75 at.% iron. In^[5], the authors observed for alloys with invar composition the simultaneous presence of a clearly pronounced hyperfine magnetic structure of the spectrum and a single line, which enabled them to conclude that they had observed the existence of two phases with ferro- and antiferromagnetic ordering of the spins. The experiments of the Japanese authors^[5,6] differed from those of Johnson et al.^[3] in that they were performed on powdered specimens of the alloys, and not on metallic foils. This difference is important because an alloy in a finely dispersed state can have a different magnetic state than in the monolithic state. We have therefore investigated the Mössbauer spectra of metallic foils of iron-nickel alloys in the entire temperature region in which magnetic order exists.

EXPERIMENT

To investigate $H_{\text{eff}}(c)$ in fcc iron-nickel alloys with the aid of nuclear γ resonance, we used alloys smelted in vacuum from pure components, with the following compositions: 11, 21, 31, 41, 46, 50, 56, 58, 61, 63, 65, 67, 69, 70, 71 at.% Fe. The deviations from the indicated compositions did not exceed 0.2 at.%.

After prolonged homogenization of the alloys (~ 100 h), samples were prepared in the form of foils rolled to a thickness 10–20 μ and quenched from 1000°C. All the alloys had an fcc structure, as established by x-ray measurements.

The spectra were measured at constant velocity, with a setup having an electrodynamic actuator from the temperature of liquid nitrogen to the Curie point of the alloy. The γ -radiation source was Co^{57} in a Pt or Pd matrix. The emission-line half-width of the source

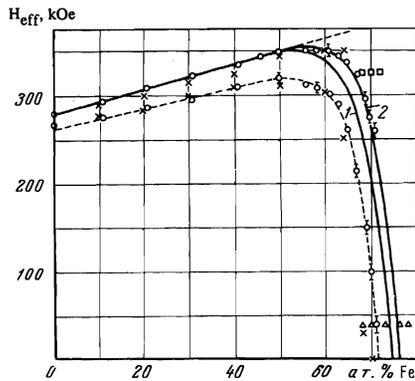


FIG. 1. Concentration dependence of the effective field at the Fe^{57} in fcc iron-nickel alloys. Experimental data: X-[²], □-[³], Δ-[⁴], ●-[⁷], ○—present work. The points near the dashed curve were obtained at room temperature, and those near the solid curve were obtained by extrapolating the temperature dependence to 0°K. Theoretical curves (solid): 1— $-\epsilon_{22} = 0$, 2— $-\epsilon_{22} = -0.02$.

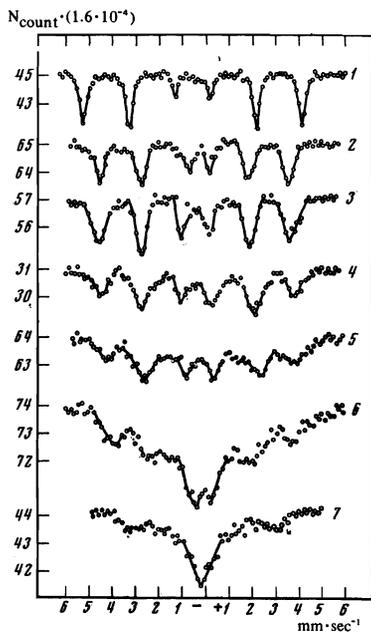


FIG. 2. Mössbauer spectra of iron-nickel alloys at the same relative temperature $\tau = 0.67$: 1—Fe (pure); 2—31% Fe + 69% Ni; 3—50% Fe + 50% Ni; 4—56% Fe + 44% Ni; 5—67% Fe + 33% Ni; 6—69% Fe + 31% Ni; 7—71% Fe + 29% Ni (the measurement error is ± 300 counts).

with Pt was approximately 1.2–1.5 of the natural half-width, and that of the Pd source was close to unity. The temperature investigations were performed in a specially constructed cryostat in nitrogen vapor and in an oven in which the temperature was automatically stabilized accurate to 0.5° .

Figure 1 shows the measured effective field at the effective field at the Fe^{57} nuclei in fcc iron-nickel alloys as the function of the composition for room temperature and for 0°K. The values of $H_{\text{eff}}(c)$ for different compositions were obtained by extrapolating to 0°K the dependence of H_{eff} on $T^{3/2}$, which was linear at low temperatures for most investigated alloys. The observed $H_{\text{eff}}(c)$ dependence shows a linear growth of the field from 0 to 50 at.% Fe, in accordance with the mixing law, and a sharp deviation from this law in the region

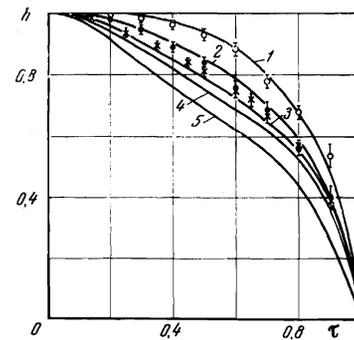


FIG. 3. Dependence of the relative field $h = H_{\text{eff}}(\tau)/H_{\text{eff}}(0)$ on the relative temperature $\tau = T/\Theta$. Solid lines—theoretical plots for iron-nickel alloys: 1—25, 2—50, 3—61, 4—65, 5—70 at.% Fe. Experimental values of $h(\tau)$: ○—31; ●—50; X—61 at.% Fe.

50–75 at.% Fe. As seen, our results coincide quite well with the results given in [³] for the compositions with 0–50 at.% Fe, and differ for the compositions with 50–75 at.% Fe. There is also a considerable disagreement with the data of the Japanese workers [^{5,6}], which is manifest not only in the fact that the numerical values of the effective magnetic fields at the Fe^{57} nuclei do not agree, but also in the form of the spectrum itself. In our case we did not observe the simultaneous presence of a sharply pronounced hyperfine magnetic structure of the spectrum and a single line in the spectra of the invar alloys.

Figure 2 shows the results of γ -resonance absorption in pure iron and in a number of alloys located both in the region of the linear section of the $H_{\text{eff}}(c)$ curve and in the region of the deviation from it. To eliminate the influence of the Curie point, which depends strongly on the concentration, the spectra are shown for the same relative temperature $\tau = T/\Theta = 0.67$. We see the absorption lines are broadened for all the compositions, and the single line noted in [⁵] for such alloys is missing from the spectra of the alloys with 67 and 69 at.% Fe. The Mössbauer spectrum for the alloy 71% Fe + 29% Ni (Fig. 2) can hardly be represented as a supposition of only two spectra. The picture is undoubtedly more complicated.

The temperature dependence of the relative field $h(\tau) = H_{\text{eff}}(\tau)/H_{\text{eff}}(0)$ is shown in Fig. 3 for some of the investigated alloys. It recalls to a considerable degree the dependence of the relative magnetization on the relative temperature τ . Both are characterized by a decrease in the values at medium τ with increasing iron concentration in the alloys.

DISCUSSION OF RESULTS

1. Magnetic Structure of Alloys

It was demonstrated in [^{4,8}] by the method of diffuse magnetic scattering of neutrons that a spatially-inhomogeneous magnetic structure exists in iron-nickel alloys in which the average magnetic moment deviates from the mixing law (invar alloys). The cause for this occurrence is the presence of antiferromagnetic interaction between the pairs of iron atoms ($J_{\text{Fe-Fe}} < 0$) in addition to the ferromagnetic interaction in the nickel-iron ($J_{\text{Ni-Fe}} > 0$) and nickel-nickel ($J_{\text{Ni-Ni}} > 0$) pairs.

Such a mixed interaction causes the z-projections of the atomic spins to become dependent on the nearest environment. Since the atoms in a disordered alloy are statistically distributed over the crystal-lattice sites, the values of these projections likewise differ from site to site. The hyperfine interaction of the nuclear spin with the electron spin of its own atom determines, in turn, the Zeeman splitting of the nuclear levels, from the value of which the effective field at the iron nuclei is estimated. These fields turn out to be significantly different at the nuclei of each of the iron atoms. Therefore the resonant-absorption peaks in the Mössbauer spectra of the investigated alloys are broadened (Fig. 2), and this broadening increases with increasing inhomogeneity of the magnetic structure, so that the spectrum for the alloys with 69 and 71 at.% Fe (spectra 6 and 7 of Fig. 2) is complicated and cannot be treated except as a superposition of a large number of spectra with different values of effective field. What is observed in experiment, on the other, is only the average value H_{eff} , obtained from measuring the distances between the centers of gravity of these lines. These values of H_{eff} are shown in Fig. 1 as functions of the concentration.

2. Description of Model and Formulas

To explain the experimentally observed $H_{\text{eff}}(c)$ dependence in the investigated alloys, it is necessary to analyze the possible causes of the appearance of an effective field at the iron nucleus. A sufficiently rigorous theory in this respect exists only for the free atom (see^[9-11]), from which it follows that the main contribution to the magnetic splitting of the nuclear levels is made by the so called Fermi "contact" interaction, which is effected via the s-electrons polarized by the 3d-electron spin. The effective Hamiltonian of such an interaction for an individual atom can be represented in the form^[11]

$$\mathcal{H} = -A_s(\mu_N \mu_s), \quad (1)$$

where μ_N and μ_s are the nuclear and electronic magnetic moments of the atom, and A_s is the constant of the hyperfine interaction of the nuclear spin with the electron spin. An analogous picture of the Zeeman splitting of nuclear levels can be obtained by assuming that the nuclear spin is situated in a certain effective field

$$H_{\text{eff}} = A_s \bar{\mu}, \quad (2)$$

where $\bar{\mu} \equiv \bar{\mu}_z$ is the mean value of the atomic magnetic moment (in the direction of the quantization axis z).

If the atom is not in the free state but in a crystal lattice, then it is necessary to take into account not only the interaction of the nuclear spin with the electron spin of the atom itself, but also with the electron spins of the neighboring atoms, and also of the atomic spins with one another. Unfortunately, the microscopic theory of these interactions in a crystal has not been sufficiently well developed. Therefore, to take into account the influence of the nearest neighbors on the effective field at the Fe^{57} nucleus, we can assume that the effective field at the nucleus is proportional to the effective molecular field at the atom (H_{mol}), which is produced by the nearest environment:

$$H_{\text{eff}} = \alpha H_{\text{mol}}, \quad (3)$$

where α is the proportionality coefficient.

For ferromagnets with perfectly identical and equivalent atoms^[12]

$$H_{\text{mol}} = \gamma \bar{\mu}_z, \quad (4)$$

where $\gamma = 2ZJ/g^2\mu_B^2$ is the constant of the molecular field, Z is the number of nearest neighbors, J is the exchange integral between two nearest atoms, g is the Landé factor, and μ_B is the Bohr magneton. Consequently, the total hyperfine field at the nucleus is

$$H_{\text{eff}} = D \bar{\mu}, \quad (5)$$

where $D \equiv \alpha\gamma$ is a certain new constant that differs from the constant of the intra-atomic hyperfine interaction A_s in that it includes the interaction of the nucleus but not only with the magnetic moment of the atom itself, but also with the atomic spins of the nearest neighbors. By itself, the interaction between the nuclear spin and the electron spins of the neighboring atoms can be also small, but the nearest environment, as already noted, changes the state of the electron spin of the atom itself, and allowance for this interaction is therefore essential for the hyperfine interaction.

We turn now to binary ferromagnetic alloys and consider, as a particular example, the iron-nickel alloys investigated by us. We label the nickel and iron atoms by the indices 1 and 2. Let c be the concentration of the iron atoms in the nickel. Then the effective molecular field at the iron atoms whose nuclear γ resonance we are investigating is written in accordance with^[8,13] in the form

$$H_{\text{mol}}(c, T) = \gamma_{21} \bar{\mu}_z^{(1)}(c, T) p_{21} + \gamma_{22} \bar{\mu}_z^{(2)}(c, T) p_{22}, \quad (6)$$

where γ_{21} and γ_{22} are the coefficients of the molecular fields produced by the atoms of type 1 and 2 at the iron atom, p_{21} and p_{22} are the corresponding probabilities of surrounding the iron atom with atoms of type 1 and 2, and $\bar{\mu}_z^{(1)}$ and $\bar{\mu}_z^{(2)}$ are the mean values of the z-projections of the atomic spins of nickel and iron, which depend in the general case on the concentration and on the temperature.

Taking into account our assumption that the effective field at the iron nucleus is proportional to the molecular field at the iron atom, we can write for the alloy

$$\bar{H}_{\text{eff}}(c, T) = D_{21} \bar{\mu}_z^{(1)} p_{21} + D_{22} \bar{\mu}_z^{(2)} p_{22}, \quad (7)$$

where D_{21} and D_{22} are certain constants which are equal identically to $\alpha\gamma_{21}$ and $\alpha\gamma_{22}$. Formula (7) goes over into formula (5) in the limiting case of pure metals. Then

$$H_{\text{eff}} = H_{21} p_{21} + H_{22} p_{22}, \quad (8)$$

where $H_{21} = D_{21} \bar{\mu}_z^{(1)}(c, T)$, $H_{22} = D_{22} \bar{\mu}_z^{(2)}(c, T)$ (9)

denote, respectively, the effective fields at the nuclei of iron atoms located in an environment of only nickel atoms or only iron atoms. Their values for pure metals can be obtained by extrapolating the linear section of the $H_{\text{eff}}(c)$ dependence to 100% Ni and Fe, or can be determined from experiments on pure metals and strongly diluted alloys. The constants D_{21} and D_{22} are assumed to be independent of the concentration and temperature, and are determined from expressions (9) if one knows the effective field at the nucleus and of the average

z-projection of the atomic spin of the iron and nickel for the given temperature and concentration. The values of $\bar{\mu}_Z^{(1)}$ and $\bar{\mu}_Z^{(2)}$ determine the average magnetic moment at this alloy in accordance with the mixing law:

$$\bar{\mu}(c, T) = \bar{\mu}_z^{(1)}(c, T)(1-c) + \bar{\mu}_z^{(2)}(c, T)c. \quad (10)$$

An analytic expression for $\bar{\mu}(c, T)$ was obtained in^[14,4] with allowance for the experimental fact that mixed ferro- and antiferromagnetic interactions exist between the atom pairs. For 0°K we have $\bar{\mu}_Z^{(1)} = \mu_1 a_1(c)$ and $\bar{\mu}_Z^{(2)}(c) = \mu_2 a_2(c)$, where $a_1(c)$ and $a_2(c)$ are the coefficients for the cancellation of the atomic spins, $\mu_1 = g\mu_B S_1$ and $\mu_2 = g\mu_B S_2$, and S_1 and S_2 are the electron spins of the nickel and iron atoms.

The temperature dependence of $H_{\text{eff}}(T)$ for a given alloy with concentration c is governed by the temperature dependence of

$$\bar{\mu}_z^{(1)}(T) = \bar{\mu}_z^{(1)}(0)\sigma_1 \text{ and } \bar{\mu}_z^{(2)}(T) = \bar{\mu}_z^{(2)}(0)\sigma_2,$$

where σ_1 and σ_2 are defined in terms of the corresponding Brillouin function:

$$\begin{aligned} \sigma_1 &= B_{S_1}(E_1/kT), & \sigma_2 &= B_{S_2}(E_2/kT), \\ E_1 &= -2ZS_1^2 J_{11} \left[a_1 p_{11} \sigma_1 + \frac{J_{12} S_2}{J_{11} S_1} p_{12} \sigma_2 \right], \\ E_2 &= -2ZS_2 S_1 J_{11} \left[\frac{J_{12}}{J_{11}} a_1 p_{21} \sigma_1 + \frac{S_2}{S_1} \frac{J_{22}}{J_{11}} p_{22} \sigma_2 \right]. \end{aligned} \quad (11)$$

Here J_{11} , J_{12} , and J_{22} are the exchange integrals between the corresponding pairs of atoms,

$$a_1(c) = (1 - Z\lambda_2 p_{22}'), \quad (12)$$

$$a_2(c) = \left[1 - Z^2 \lambda_1 \lambda_2 \frac{c}{1-c} p_{22}'(1-p_{22}') \right],$$

Z is the number of nearest neighbors, λ_1 and λ_2 are certain constants whose calculation is described in^[14],

$$p_{22}' = \frac{1}{Z} \left[\sum_0^Z P_z(n, p_{22})n - \sum_0^{X-1} P_z(n, p_{22})n \right], \quad (13)$$

where $X = 1, 2, \dots, Z$, and $P_z(n, p_{22})$ satisfies the binomial distribution

$$P_z(n, p_{22}) = \frac{Z!}{n!(Z-n)!} p_{22}^n (1-p_{22})^{Z-n}. \quad (14)$$

For disordered alloys $p_{22} = c$, and taking into account the atomic correlation (cf.^[15]) we have $p_{22} = c + \epsilon_{22}/c$, where ϵ_{22} is the correlation parameter on the first coordination sphere. For alloys with ordering of the Ni_3Fe type, with a degree of long-range order η_* we have

$$p_{22} = c - \gamma^2 \eta_*^2 / 9c, \quad (15)$$

with $\gamma = c(1-\nu)/\nu$ for $c \leq 1/4 = \nu$ and $\gamma = 1-c$ for $c \geq 1/4$. For alloys with ordering of the NiFe type

$$p_{22} = c - \gamma^2 \eta_*^2 / 3c, \quad (16)$$

where $\gamma = c(1-\nu)/\nu$ for $c \leq \nu = 1/2$ and $\gamma = 1-c$ for $c \geq 1/2$. Thus, the temperature dependence of the reduced field at the iron nucleus is determined for the given alloy by the expression

$$h(\tau) = \frac{D_{21} \bar{\mu}_z^{(1)}(0) \sigma_1 p_{21} + D_{22} \bar{\mu}_z^{(2)}(0) \sigma_2 p_{22}}{D_{21} \bar{\mu}_z^{(1)}(0) p_{21} + D_{22} \bar{\mu}_z^{(2)}(0) p_{22}}. \quad (17)$$

Taking (15) and (16) into account, formula (7) enables us also to calculate $H_{\text{eff}}(c)$ in both disordered and ordered states.

3. Comparison with Experiment

For a comparison with experiment, it is necessary to know the two parameters D_{21} and D_{22} in (7). The quantities are obtained from (9) if one determines the magnetic fields at Fe^{57} nuclei having only a nickel environment (H_{21}) or only an iron environment (H_{22}) in an fcc lattice. The values of H_{21} and H_{22} are determined from the experimental $H_{\text{eff}}(c)$ dependence by continuing the linear section of the curve to pure nickel and iron. From the data shown in Fig. 1 we obtain $H_{21} = 280$ kOe and $H_{22} = 420$ kOe. Recognizing that in the composition region 0–50 at.% Fe the z-projection of the atomic spins of nickel and iron coincide with their total values $\mu_{\text{Fe}} = 2.8 \mu_B$ and $\mu_{\text{Ni}} = 0.6 \mu_B$ obtained from neutron-diffraction data^[4], we obtain the following numerical values of the constants for fcc lattices: $D_{21} = 5.3 \times 10^{24}$ G²/erg and $D_{22} = 1.62 \times 10^{24}$ G²/erg. The constants D_{21} and D_{22} are universal to some degree. This is seen from the fact that D_{22} turns out to be exactly the same when calculated from the data on the effective magnetic field for pure iron in the fcc lattice ($H_{\text{eff}} = 339$ kOe, $\mu_{\text{Fe}} = 2.2 \mu_B$, $D_{22} = 1.62 \times 10^{24}$ G²/erg). This agreement is not accidental, but is a reflection of the fact that the interaction of the nuclear spin of the iron atom with the electron spin of the neighboring atom of the same sort via the electron spin of the atom itself are the same for ferromagnetic iron with bcc and fcc lattices. Indeed, formula (9) contains no dependence on the type of lattice.

The solid curves of Fig. 1 were calculated from formula (7). Curve 1 pertains to perfectly disordered alloys, and curve 2 to alloys having a short-range atomic order. The correlation $\epsilon_{22} = -0.02$, assumed in the calculation, agrees with the experimentally measured value ϵ_{22} in these alloys^[16]. We see that when the atomic correlation is taken into account the agreement between calculation and experiment is good.

To calculate the temperature dependence of the effective field at the Fe^{57} nuclei in alloys, it is necessary to know the $\sigma_1(\tau)$ and $\sigma_2(\tau)$ dependences. They were obtained assuming the spin values $S_1 = 1/2$ and $S_2 = 3/2$ and the exchange-integral ratios $J_{12}/J_{11} = 0.92$ and $J_{22}/J_{11} = 0.05$. For these values of the spins and exchange integrals, good agreement was observed between the theoretical and experimental dependences of the Curie point on the concentration and of the saturation magnetization on the temperature for all iron-nickel alloys with fcc lattice^[13]. The $h(\tau)$ curves calculated in the same approximation are shown in Fig. 3 by solid lines. Satisfactory agreement with experiment is observed, and, most importantly, it becomes possible to explain the evolution of the $h(\tau)$ curve with increasing iron concentration in the nickel.

Formula (7) makes it also possible to explain the difference between the values of the effective magnetic fields at the Fe^{57} nuclei in disordered and ordered states of the alloys. It was shown in^[17,18] that H_{eff} in the stoichiometric Ni_3Fe alloy is larger in the disordered state than in the ordered state. In accordance with formula (9), the ordered Ni_3Fe ($\eta_* = 1$) has no atoms of the same sort in the nearest surroundings of the iron atoms, and this leads to a decrease of the effective field at the iron nucleus, from 315 kOe (according to our

data) to 278 kOe according to^[17]. An analogous decrease of the field can be observed also for alloys with ordering of the NiFe type. At the same time, in the composition range from 55 to 75 at.% Fe, an increase of the degree of order leads to an increase in the field at the nucleus. This is caused to a considerable degree by a change of the z-projections of the atomic spins of iron and of the nickel following the ordering in the alloys of these compositions.

CONCLUSION

Thus, our detailed investigations of fcc iron-nickel alloys with the aid of the Mössbauer effect have made it possible to confirm the earlier conclusion^[4] that a spatially-inhomogeneous magnetic structure exists in invar alloys. This conclusion follows from the form of the Mössbauer spectra obtained with monolithic alloy foils. The latter circumstance is very important, since measurements on finely-dispersed powders reflect apparently a somewhat different state of the alloy, and therefore the conclusions from such investigations can not be extended without stipulation to the magnetic state in monolithic alloys. Using the previously developed notions concerning the magnetic structure and assuming proportionality of the effective field at the iron nucleus to the molecular field, which produce the nearest-environment atoms at the given atom, we were able to explain the concentration dependence of the field at the Fe⁵⁷ nucleus, and also the difference between the average effective fields at the iron nuclei in alloys in ordered and disordered states. This approach explains also the temperature dependence of the field at the nucleus for different alloys of the iron-nickel system with fcc lattice. We also assume that the approach developed here can be fruitful for other two-component iron alloys.

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