

# Concentration dependence of the magnetization and the Curie temperature in disordered Fe–M alloys (M=Al, Si, Sn)

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The results of theoretical and experimental investigations of the temperature and concentration dependences of the magnetic properties of disordered Fe–M alloys (M=Al, Si, Sn) at low concentrations of the metalloid atoms are presented. The concentration dependences of the magnetic moment and the Curie temperature, whose behavior accounts for the main laws governing the experimental results, are calculated using functional integration. It is shown that the use of localized models to describe the thermodynamic properties of the alloys investigated is incorrect, since even in the absence of Stoner excitations the thermodynamic fluctuations of the electron and spin densities renormalize the effective exchange interaction. This renormalization is different at different concentrations of the metalloid atoms. © 1996 American Institute of Physics. [S1063-7761(96)02709-6]

## 1. INTRODUCTION

A fairly large amount of experimental material on disordered binary alloys of iron with nonmagnetic *sp* elements, such as Al and Si, has accumulated in the last few years. It includes the results of crystallographic and Mössbauer investigations, investigations of the concentration and temperature dependences of the magnetization and susceptibility, as well as of the temperature of the phase transition from the magnetically ordered state to the disordered state.<sup>1–10</sup> The possibility of obtaining an atomically disordered state over broad concentration ranges makes these alloys good model systems for studying the fundamental laws governing the shaping of magnetic properties. The general features of the laws observed point to a limited number of factors that determine them. In fact, when it was assumed only that the *d* states of the metalloid atoms are at large distances along the energy scale from the Fe *d* band and that the *s*–*d* hybridization in an Fe atom is determined by the number of metalloid atoms in the local environment, the main laws governing the shaping of the magnetism of these alloys could be described qualitatively.<sup>11,12</sup> It was also found that there are significant manifestations of the band character of the magnetism at high impurity concentrations. They include a decrease in the amplitudes of the local magnetic moments with the concentration and the presence of Stoner-type excitations at low temperatures, which cause both changes in the magnitude of the individual local magnetic moments and changes in their directions. It therefore became clear that the description of the magnetic states of these alloys on the basis of localized models like the Ising and Heisenberg models<sup>13,14</sup> is not justified. However, at low impurity concentrations, at which the magnitude of the local magnetic moment scarcely varies with the concentration and the Stoner-type excitations are insignificant, this assertion is in need of additional substantiation.

In this paper we concentrate our attention on experimental and theoretical investigations of unorderd Fe–M alloys (M=Al, Si, Sn) at concentrations of the metalloid atoms below 10 at. %.

## 2. EXPERIMENTAL RESULTS

As was noted above, a fairly large amount of experimental material on unorderd alloys has been accumulated. Figures 1–3 present published data,<sup>3–10,15–19</sup> some of which were previously obtained by one of us.<sup>9,10,19</sup> The samples used in Refs. 9, 10, and 19 were obtained by mechanical grinding similar to that described below for the Fe–Sn alloys. Mechanical treatment in a planetary ball mill ensures the formation of homogeneous disordered crystalline alloys<sup>1,2</sup> and substitutional alloys at room temperature.

Fe–Sn alloys with Sn concentrations equal to 3.2, 6.0, and 7.6 at. % were smelted in an argon induction furnace from the high-purity components Fe (99.95) and Sn (99.99). The ingots obtained were ground for 20 h in a Pulverizette planetary ball mill in an argon atmosphere at room temperature. Tungsten carbide (WC) grinding vessels and balls, which ensure minimal contamination of the ground samples by the material of the vessels and the balls as a result of wear, were used. According to the data from chemical analysis, the maximum WC content was equal to 5% by weight for the sample with 3.2% by number of Sn, and in the other samples its content did not exceed 1% by weight. The mean diameter of the particles after grinding, which was evaluated on an Analizette-22 laser interferometer, was equal to 4  $\mu\text{m}$ .

The structural state of the alloys was monitored by x-ray diffraction analysis and Mössbauer spectroscopy. The x-ray diffraction pattern of the alloy with 3.2% by number. Sn displayed low-intensity lines from WC along with intense bcc reflections. The diffraction patterns of the alloys with

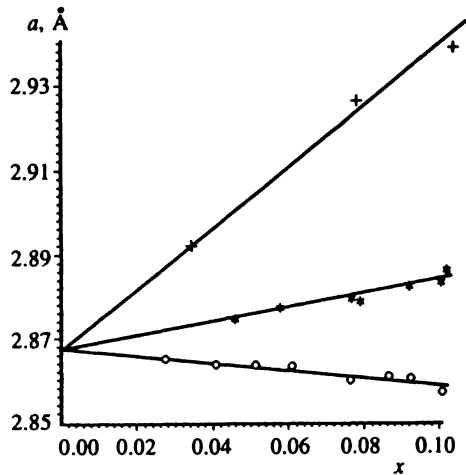


FIG. 1. Concentration dependence of the bcc lattice parameter  $a$  of unordered alloys: (\*)—Fe—Al (Refs. 10 and 18), (O)—Fe—Si (Refs. 16, 17, and 19), (+)—Fe—Sn (present work).

6.0% and 7.6% Sn by number contained only bcc reflections. No lines from the hexagonal intermetallic compound  $\text{Fe}_3\text{Sn}$ , which might be present in the samples according to the equilibrium phase diagram,<sup>20</sup> were discovered. An analysis of the components of the magnetically split Mössbauer spectra revealed a random distribution of the Fe and Sn atoms among the sites of the bcc lattice.

The Curie temperature of the bcc disordered Fe—Sn alloys was determined from the temperature dependence of the magnetic susceptibility measured in a variable sinusoidal field with an amplitude of 80 A/m and a frequency of 120 Hz. For the measurements the sample was placed in a quartz ampoule filled with argon. The junctions of the thermocouple were introduced directly into the powdered sample, making it possible to use high heating rates to traverse the two-phase region on the equilibrium phase diagram to the temperature range with the equilibrium bcc phase, in which the sample was held for 1 h. Then the sample was cooled or heated relatively slowly in the vicinity of the Curie temperature.

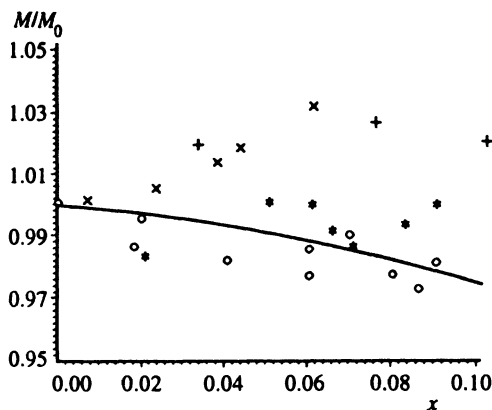


FIG. 2. Concentration dependence of the mean magnetic moment in an Fe atom in disordered alloys: (\*)—Fe—Al (Refs. 3, 4, and 15), (O)—Fe—Si (Refs. 3, 4, and 9), (X)—Fe—Sn (Ref. 3), (+)—Fe—Sn (present work), solid line—calculation in the present work.

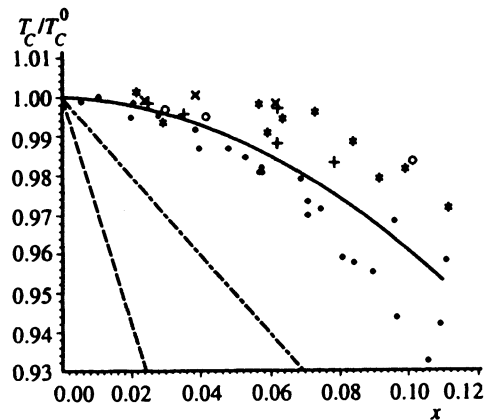


FIG. 3. Concentration dependence of the Curie temperature of unordered alloys: (\*)—Fe—Al (Refs. 3, 4, 8, and 10), (O)—Fe—Si (Refs. 3–7 and 9), (●)—Fe—Ga (Ref. 8), (X)—Fe—Sn (Ref. 3), (+)—Fe—Sn (present work), dot-dashed line—Heisenberg model, dashed line— $T_c(x) \sim J_{\text{eff}}(x)$ , solid line—calculation in the present work.

It is noteworthy that the literature data and the data from our experiments were selected in a special manner. They contain metalloid atoms with  $p$  subshells having different electronic configurations (Al— $3p^1$ , Ga— $4p^1$ , Si— $3p^2$ , Sn— $5p^2$ ) and different covalent radii (Al—1.18 Å, Ga—1.26 Å, Si—1.11 Å, Sn—1.41 Å). Nevertheless, the magnetic properties of these alloys (see Figs. 2 and 3) differ weakly from one another.

The existing differences between the concentration dependences are not stipulated by the distances between the atoms (Fig. 1) or by the electronic configuration of the metalloid atoms.

### 3. SUBSTANTIATION OF THE MODEL HAMILTONIAN. CALCULATION OF THE CONCENTRATION DEPENDENCES OF THE MAGNETIZATION AND THE CURIE TEMPERATURE

In the 1980s, a large number of theoretical studies were devoted to devising the theory of spin fluctuations.<sup>21–26</sup> This theory was devised at once as an interpolation between two diametrically opposed limits: band and localized magnetism. Therefore, it explains the main laws governing the magnetic properties of transition metals relatively well. It is natural to use it in our case, too. We have several arguments for doing so, which have already been partially noted in the preceding section. First, the differences in the concentration dependences for different metalloid atoms (Figs. 2 and 3) are small, despite the existence of differences in the electronic configurations and radii of the atoms. This allows us to assume that these differences are not among the factors that determine the magnetic properties. Second, these alloys can be regarded as unordered substitutional alloys with a good degree of accuracy. Finally, the low concentrations of the metalloid atoms allow us to neglect the changes in the  $sp-d$  hybridization, which are significant at high concentrations.<sup>11,12</sup> Therefore, we can assume that the  $sp$  electrons of the iron and metalloid atoms form a broad com-

mon band and that the  $d$  states, which are at a large distance along the energy scale, do not interact with them. We write the Hamiltonian in the following form:

$$H = \sum_{i\sigma} \varepsilon_i c_{i\sigma}^+ c_{i\sigma} + \sum_{ij} t_{ij} c_{i\sigma}^+ c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} \\ = H_0 + H_{\text{int}}, \\ H_{\text{int}} = \sum_{i\sigma} \varepsilon_i c_{i\sigma}^+ c_{i\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}. \quad (1)$$

Here the relation between the strengths of the direct and exchange interactions is taken into account;<sup>23</sup>  $\varepsilon_i$  takes the value  $\varepsilon^A$  for the metalloid atoms and  $\varepsilon^B$  for the iron atoms, where  $|(\varepsilon^A - \varepsilon^B)/W| \gg 1$ ;  $c_{i\sigma}^+$  ( $c_{i\sigma}$ ) is the creation (annihilation) operator of an electron with spin  $\sigma$  at site  $i$ ;  $n_{i\sigma} = c_{i\sigma}^+ c_{i\sigma}$ ; and  $U$  is the intraatomic electron interaction constant. Leaving the values of the  $t_{ij}$  and  $U$  for the two atoms identical (corresponding to Fe) does not introduce a significant error, since the  $d$  band of the metalloid is at a considerable distance from the Fermi level and remains unfilled. The Hamiltonian (1) is single-band. As was shown in Ref. 23, calculations in the single-band model followed by renormalization of the free energy and the magnetic moment take into account both the degeneracy with respect to the orbital quantum number and Hund's rule. When  $\varepsilon_i = \text{const}$ , the Hamiltonian completely coincides with the Hamiltonians previously used to describe the properties of pure transition metals. This points out the possibility of using functional integration<sup>27</sup> in our case.

Using the equality

$$\sum_i n_{i\uparrow} n_{i\downarrow} = \sum_i \left[ \frac{n_i^2}{4} - S_{iz}^2 \right], \quad (2)$$

where  $n_i = n_{i\uparrow} + n_{i\downarrow}$  and  $S_{iz} = (n_{i\uparrow} - n_{i\downarrow})/2$ , we write the partition function of the system in the interaction representation:<sup>28</sup>

$$Z = \text{Tr} \left\{ \exp(-\beta \tilde{H}_0) T_\tau \exp \left[ - \int_0^\beta d\tau H_{\text{int}}(\tau) \right] \right\}, \quad (3)$$

where  $\tilde{H}_0 = H_0 - \mu N$ ,  $\mu$  is the chemical potential,  $N$  is the total-number-of-electrons operator,  $\beta = 1/T$  is the reciprocal temperature in energy units, and  $T_\tau$  is the ordering operator with respect to the imaginary time  $\tau$ . Here and in the following the operators in the interaction representation are

$$A(\tau) = \exp[\tau \tilde{H}_0] A \exp[-\tau \tilde{H}_0].$$

Using the Stratonovich-Hubbard identity, we go over to the functional integral:<sup>22</sup>

$$Z = \int \left( \prod_i \delta x_i(\tau) \delta v_i(\tau) \right) \exp \left[ - \frac{\pi}{\beta} \int_0^\beta d\tau \sum_i [x_i^2(\tau) + v_i^2(\tau)] \right] Z\{x_i(\tau), v_i(\tau)\}, \quad (4)$$

$$Z\{x_i(\tau), v_i(\tau)\} = \text{Tr} \left\{ \exp(-\beta \tilde{H}_0) T_\tau \right. \\ \times \exp \left[ - \frac{1}{\beta} \int_0^\beta d\tau \sum_i \left[ 2\sqrt{\pi\beta U} v_i(\tau) S_z(\tau) + \sqrt{\pi\beta U} i x_i(\tau) n_i(\tau) + \beta \varepsilon_i n_i(\tau) \right] \right] \right\}. \quad (5)$$

This transition is possible, since all the operators in  $H_{\text{int}}$  commute with one another, including the term associated with disorder. Introducing the site-specific spin density matrix  $\rho_{i,\sigma\sigma'}(\tau) = a_{i\sigma}^+(\tau) a_{i\sigma}(\tau)$ , we can represent the latter expression in the form

$$Z\{x_i(\tau), v_i(\tau)\} = \text{Tr} \exp(-\beta \tilde{H}_0) T_\tau \\ \times \exp \left\{ - \int_0^\beta d\tau \sum_l \text{Tr}_\sigma (V_l(\tau) \rho_l(\tau)) \right\}, \quad (6)$$

where the potential  $V_l(\tau)$  at site  $l$  is a diagonal matrix, in which, besides the fluctuating fields,  $\varepsilon_l$  varies as a function of the type of site:

$$V_l(\tau) = \begin{pmatrix} \sqrt{\frac{\pi U}{\beta}} [i x_l(\tau) + v_l(\tau)] + \varepsilon_l & 0 \\ 0 & \sqrt{\frac{\pi U}{\beta}} [i x_l(\tau) - v_l(\tau)] + \varepsilon_l \end{pmatrix}.$$

With the exception of  $\varepsilon_l$ , Eq. (6) is the standard expression for representing the Hubbard model by a two-field functional integral.

We shall not discuss the approximations made here in detail. There are numerous papers in which they were substantiated, for example, Refs. 21-26. In addition, there is presently no unified opinion regarding particular approximations, and they must be analyzed in each specific case. On the basis of the formulation of our problem and its purposes (determination of the concentration dependence of the magnetic characteristics), it seems perfectly justified to us to apply the static single-site approximation for spin fluctuations<sup>21,22</sup> and to disregard the charge fluctuations, which are rapid variables relative to the spin fluctuations, after replacing them by potentials chosen according to the condition that the site be electroneutral.<sup>24</sup> In this approximation

$$Z = Z_0 \int \left( \prod_l dv_l \right) \exp \left\{ - \pi \sum_l v_l^2 - \frac{\beta}{\pi} \int_{-\infty}^{+\infty} dz \frac{\text{Tr} \text{Im} \ln(1 - G^0 V)}{e^{(z-\mu)/T} + 1} \right\}. \quad (7)$$

Here  $G^0$  and  $V$  are the matrices at sites  $l$  and  $l'$ , which consist of the  $2 \times 2$  blocks

$$V_{ll'} = \begin{pmatrix} \sqrt{\frac{\pi U}{\beta}}(w_l + v_l) + \varepsilon_l & 0 \\ 0 & \sqrt{\frac{\pi U}{\beta}}(w_l - v_l) + \varepsilon_l \end{pmatrix} \Delta_{ll'} = \tilde{V}_l,$$

where  $\Delta_{ll'}$  is the Kronecker delta, and

$$G_{ll'}^0 = \begin{pmatrix} ((z - H_0)^{-1})_{ll'} & 0 \\ 0 & ((z - H_0)^{-1})_{ll'} \end{pmatrix}.$$

It was shown in Ref. 12 that at low impurity concentrations the Stoner excitations in these alloys are insignificant; therefore, the Fermi distribution function in (7) was replaced by a step function.

Using the single-site approximation of the coherent potential,<sup>22</sup> we replace  $\text{Tr} \ln(1 - G^0 V)$  by  $\Sigma_l \ln[1 - g(\tilde{V}_l - \Sigma)]$ , where  $g$  and  $\Sigma$  are  $2 \times 2$  matrices in the spin variables

$$\Sigma(z) = \begin{pmatrix} \Sigma^\uparrow(z) & 0 \\ 0 & \Sigma^\downarrow(z) \end{pmatrix},$$

$$g = \begin{pmatrix} g^\uparrow & 0 \\ 0 & g^\downarrow(z) \end{pmatrix},$$

$$g^\sigma = (z - H_0 - \Sigma^\sigma(z))_{ll}^{-1}.$$

The equation for the self-energy parts  $\Sigma^\uparrow(z)$  and  $\Sigma^\downarrow(z)$  is obtained from the condition that the averaged single-site scattering matrix be equal to zero:

$$\int_{-\infty}^{+\infty} dv \int_{-\infty}^{+\infty} d\varepsilon_l t_l W(v, \varepsilon_l) = 0. \quad (8)$$

Here  $t_l = (\tilde{V}_l - \Sigma)[1 - g(\tilde{V}_l - \Sigma)]^{-1}$ , and  $W(v, \varepsilon_l)$  is the distribution probability density of  $v = \sqrt{\pi U T} v_l$  and  $\varepsilon_l$  at site  $l$ :

$$W(v, \varepsilon_l) = C [x \delta(\varepsilon^A - \varepsilon_l) + (1 - x) \delta(\varepsilon^B - \varepsilon_l)] \times \exp \left\{ -\frac{1}{UT} \left( v^2 + \frac{U}{\pi} \int_{-\infty}^{\mu} dz \text{Tr}_\sigma \text{Im} \ln [1 - g(\tilde{V}_l - \Sigma)] \right) \right\}, \quad (9)$$

where  $C$  is a normalization constant. The equality (8) together with the condition for the chemical potential  $\mu$

$$N_{el} = -\frac{1}{\pi} \int_{-\infty}^{\mu} dz \sum_{\sigma} \text{Im} g^{\sigma}(z) \quad (10)$$

form a closed system of equations. A seed density of states (Fig. 4), which models the density of states for Fe, was used for the calculations. The parameters of the model density of states, the number of  $d$  electrons below the Fermi level  $N_{el} = 1.485$ , and  $U = 1.1$  were selected such that they and the physical parameters (the magnetic moment  $M_0$  in an Fe atom at  $T = 0$  and  $x = 0$ ;  $T_C^0 = T_C$  at  $x = 0$ ) obtained when they are used would be close to the experimentally observed values. Similar parameters and a similar model density of states have been used repeatedly with little variation of their values. Our model density of states corresponds to Ref. 26.

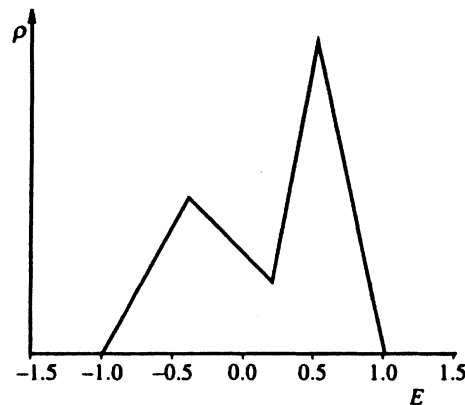


FIG. 4. Dependence of the seed density of states  $\rho$  on the energy (the energy units are adjusted to the half-width of the band).

We should not expect absolute agreement for  $M_0$  and  $T_C^0$  under the approximations that we made. Therefore, to avoid any misperceptions, we present calculated data converted to  $M_0$  and  $T_C^0$ . The magnetic moment in an Fe atom was calculated using the familiar formula

$$M = -\frac{1}{\pi} \int_{-\infty}^{\mu} dz (\text{Im} g^\downarrow(z) - \text{Im} g^\uparrow(z)).$$

The Curie temperature  $T_C$  was set equal to the temperature at which the magnetic moment vanishes. The concentration dependences thus obtained are presented in Figs. 2 and 3. Figure 3 also presents the dependence

$$J_{\text{eff}}(x) = \int_{-\infty}^{\mu} dz \text{Tr}_\sigma \text{Im} \ln [1 - g(\tilde{V}_l - \Sigma)] \Big|_{v = MU/2, \varepsilon_l = \varepsilon^B} - \int_{-\infty}^{\mu} dz \text{Tr}_\sigma \text{Im} \ln [1 - g(\tilde{V}_l - \Sigma)] \Big|_{v = -MU/2, \varepsilon_l = \varepsilon^B}. \quad (11)$$

This is the difference between the free energies of states with oppositely oriented spins on an isolated Fe site at  $T = 0$ . According to its meaning, it is an effective exchange interaction, from whose concentration dependence  $T_C$  is most often extrapolated under the assumption that  $J_{\text{eff}}(x) \sim T_C(x)$ . It should be noted that at  $T = 0$  the solution of Eqs. (8) and (10) naturally leads to the Hartree-Fock approximation and the coherent potential approximation for the diagonal disorder of the random quantity  $\varepsilon_l$ . This is easily seen by going to the limit  $T \rightarrow 0$  in (9):

$$W(v, \varepsilon_l) \rightarrow (x \delta(\varepsilon^A - \varepsilon_l) + (1 - x) \delta(\varepsilon^B - \varepsilon_l)) \delta \left( v - \frac{MU}{2} \right).$$

#### 4. CONCLUSIONS

Thus, the samples of unordered Fe-Sn alloys obtained and the experimental investigations performed on them together with the experimental data in the literature on unordered Fe-Al, Fe-Si, Fe-Ga, and Fe-Sn bcc alloys make up a set which has enabled us to formulate a model Hamiltonian for describing the thermodynamic properties of these alloys

at low concentrations of the metalloid atoms. Using this Hamiltonian, we were able to describe the main laws governing the concentration dependences of the Curie temperature and the magnetization of these alloys at low temperatures within the theory of spin fluctuations for band magnets. Depending on the metalloid atoms, a more accurate description can probably be obtained by taking into account the  $sp-d$  hybridization. This requires departure from the single-site approximation and consideration of the second band. As was noted above, such calculations were performed in the ground state in Refs. 11 and 29. Using them, we can explain the slight increase in the magnetic moment in the Fe–Sn system (Fig. 2). In addition, they predict an increase in the number of  $d$  electrons below the Fermi level as the concentration of the metalloid atoms increases. As our calculations show, an increase in the number of  $d$  electrons leads to a decrease in the slope of the plot of  $T_C(x)$ . Thus, the competition between two mechanisms, viz., the increase in the number of  $d$  electrons and the flattening of the  $d$  band due to  $s-d$  hybridization, determine the detailed dependence of  $T_C$  on  $x$ .

A second important conclusion that can be drawn from this work is as follows. Despite the fact that the local magnetic moment remains constant and the Stoner fluctuations are negligibly small, localized models (like the Ising and Heisenberg models that were used in Refs. 13 and 14) cannot be used to describe the thermodynamic magnetic properties of these alloys. For example, Fig. 3 presents plots of  $T_C(x)$  in the single-site approximation for the Heisenberg model with nonmagnetic impurities<sup>30</sup> and the extrapolation  $T_C(x) \sim J_{\text{eff}}(x)$ , where  $J_{\text{eff}}(x)$  is the effective exchange interaction calculated from Eq. (11) at  $T=0$ . Both plots are clearly far from the experimentally observed dependences. This is because  $J_{\text{eff}}(x)$  varies as the temperature increases due to the spin-density fluctuations and their influence is different at different concentrations of the metalloid atoms.

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