

Magnetic properties of Fe–Ni invar calculated in the dynamic non-local approximation of the spin fluctuation theory

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

2004 J. Phys.: Condens. Matter 16 361

(<http://iopscience.iop.org/0953-8984/16/3/015>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 129.252.86.83

The article was downloaded on 28/05/2010 at 07:50

Please note that [terms and conditions apply](#).

Magnetic properties of Fe–Ni invar calculated in the dynamic non-local approximation of the spin fluctuation theory

B I Reser

Institute of Metal Physics, Ural Branch of the Russian Academy of Sciences,
620219 Ekaterinburg, GSP-170, Russia

E-mail: reser@imp.uran.ru

Received 30 April 2003

Published 9 January 2004

Online at stacks.iop.org/JPhysCM/16/361 (DOI: 10.1088/0953-8984/16/3/015)

Abstract

The magnetization, the Curie temperature, the local magnetic moment and susceptibilities of Fe–Ni invar are calculated, using the first-principles density of states and the dynamic non-local approximation of the spin-fluctuation theory. A good agreement with experimental data over a wide range of temperatures, including low temperatures, has been found.

1. Introduction

The invar problem has been the subject of many experimental and theoretical investigations performed in the last decades (for a review see [1–4]). However, no general agreement on the origin of the invar effect has been reached so far. One of the most commonly accepted models is the 2γ -state model [5], which is supported by several band structure and total energy calculations [6–8] and experimental investigations (see [9] and references therein). But in recent papers [10–12] it is claimed that the interpretation of the invar effect on the basis of the two-state model is incorrect. In [10, 11] this conclusion is drawn from the noncollinear calculations of the ground-state properties of Fe–Ni invar at different volumes, in [12] from the polarized neutron diffraction measurements. Today there is no direct experimental evidence for the existence of high-moment and low-moment states for Fe in the Fe–Ni invar, and above all, it is difficult to justify this hypothesis on the basis of the modern theory of metallic magnetism (see, e.g. [13, 14]).

The modern approach to the invar problem, as well as to other problems connected with thermodynamics of magnetic transition metals, is based on the spin-fluctuation theory (SFT). The development of this theory goes in two directions. The first direction, that originates from the paper [15], is the *phenomenological* SFT based on the Ginzburg–Landau expansion of the free energy. For invars this approach has been developed in [16] and applied to Fe–Ni alloy in the paper [17]. Unfortunately, the approach [15, 16] has been developed for

weak itinerant ferromagnets [18] only and its suitability for Fe–Ni invar with large magnetic moment and large Curie temperature is not well founded. The second direction is the single-site approximation [19, 20] of the SFT applied to Fe–Ni alloys in [21, 22]. The calculations [21, 22] are microscopic and based on the exact functional-integral method [23, 24]. However, in the calculations [21, 22] a number of significant simplifications were made. In particular, the initial density of states (DOS) was chosen in a simplified model form, and the two-field model of spin fluctuations was used, which means a neglect of the vectorial nature of magnetic moment. Most important, the calculations [21, 22] were performed in the *static* approximation, i.e. the spin fluctuations were treated classically, while as is known the static approximation overestimates the contribution due to spin fluctuations and yields an incorrect temperature dependence of the magnetization, especially at low temperatures.

In the present paper, for the first time we use the dynamic non-local approximation (DNA) of the SFT, developed in [25] and successfully applied to Fe, Co and Ni in [26–28], for investigation of the temperature dependence of the magnetic properties of Fe–Ni invar. Our approach is based on a self-consistent quadratic approximation for the free energy of electrons in a random exchange field. The initial parameters of the calculation are the DOS and the magnetic moment per atom at $T = 0$ for Fe–Ni invar.

2. Theoretical model

A method for self-consistent calculation of magnetic properties of ferromagnetic metals at finite temperatures, based on the usage of real band structure and the DNA of the SFT, was developed in [25]. Let us give the basic ideas of the method and present the final formulae essential for an understanding of the results of our investigation.

Using Stratonovich–Hubbard transformation [23, 24], the pair interaction of electrons characterized by the intratomic electron repulsion constant U is replaced by the interaction of electrons with the exchange field $V \equiv (V_1, V_2, \dots)$, $V_j = \mathbf{V}_j(\tau)\boldsymbol{\tau}$, fluctuating in space and in ‘time’ $\tau \in [0, 1/T]$, where j is the site number, the $\boldsymbol{\tau}$ are the Pauli matrices and T is the temperature in energy units. (We neglect rapid charge fluctuations.) After this transformation the grand partition function is written as

$$\Xi = \int e^{-\Omega_0(V)/T} e^{-\Omega_1(V)/T} \prod_j D\mathbf{V}_j(\tau) \bigg/ \int e^{-\Omega_0(V)/T} \prod_j D\mathbf{V}_j(\tau) \quad (1)$$

where

$$\Omega_0(V) = \frac{1}{2} \text{Tr}(V^2/U) \quad (2)$$

is the energy of the random exchange field and

$$\Omega_1(V) = T \text{Tr} \ln G(V) = T \text{Tr} \ln[G^0(1 - VG^0)^{-1}] \quad (3)$$

is the grand potential expressed in terms of the single-particle Green function for electrons in an external field V

$$G(V) = (z + \mu - H_0 - V)^{-1}. \quad (4)$$

Here z is the energetic variable, which may take complex values; μ is the chemical potential; H_0 is the operator of the energy of the band electrons. Tr means summation of all diagonal elements, the result of which is independent of the choice of the specific matrix representation. The zeroth Green function G^0 is defined by (4) with $V = 0$.

The matrices V and G^0 cannot be diagonalized simultaneously either in coordinate or in momentum spaces. For this reason, the formal exact expression (3) is inconvenient in calculations and should be simplified.

First we turn to the Helmholtz free energy of the electrons in a fluctuating field, $F(V) = \Omega(V) + \mu N_e$, where $\Omega(V) = \Omega_0(V) + \Omega_1(V)$ and N_e is the total number of electrons. Taking into account (2) and (3), we have

$$F(V) = T \operatorname{Tr} \left(\frac{V^2}{2UT} + \ln G(V) \right) + \mu N_e. \quad (5)$$

Now we approximate this function by a quadratic form. For this purpose, we expand $F(V) = F(\langle V \rangle + \Delta V)$ in powers of the fluctuations ΔV to the second order and replace the corresponding derivatives by their mean values [29], which leads to the closest quadratic approximation

$$F(V) \simeq \sum_{\mathbf{q}n\alpha} \Delta V_{\mathbf{q}n}^\alpha (U^{-1} - \chi_{\mathbf{q}n}^\alpha) \Delta V_{-\mathbf{q}-n}^\alpha \quad (6)$$

where

$$\chi_{\mathbf{q}n}^\alpha \equiv \chi_{\mathbf{q}}^\alpha(i\omega_n) = -\frac{1}{2} T \operatorname{Sp} \sum_{\mathbf{k}m} \langle G(V) \rangle_{\mathbf{k}m} \tau^\alpha \langle G(V) \rangle_{\mathbf{k}-\mathbf{q}, m-n} \tau^\alpha \quad (7)$$

is the unenhanced dynamic susceptibility (in units of $g^2 \mu_B^2 / 2$). The fluctuation of the exchange field ΔV^α ($\alpha = x, y, z$) and the Green function $G(V)$ are written down here in the momentum–‘frequency’ ($\mathbf{q}\omega_n$) representation. Sp means the sum of diagonal elements over the spin projection index $\sigma = \uparrow, \downarrow$ or ± 1 and the mean is defined by the formulae

$$\langle \dots \rangle = \int c(V) \dots d\mathbf{V} \quad c(V) = \exp(-F(V)/T) / \int \exp(-F(V)/T) d\mathbf{V}. \quad (8)$$

Note that in the derivation of formulae (6) and (7), the expansion of the free energy $F(\langle V \rangle + \Delta V)$ is taken at $\langle V \rangle$ determined from the condition that the linear term vanishes. In the quadratic term, the Green function $G(V)$ is replaced by $\langle G(V) \rangle$. The unimportant constant value $\langle F(V) \rangle$ is discarded in the expansion.

We represent Green function (4) as

$$G(V) = G(\Sigma) [1 - (\Delta V - \Delta \Sigma) G(\Sigma)]^{-1} \quad (9)$$

where

$$G(\Sigma) = (z - H_0 - \Sigma)^{-1} \quad (10)$$

is the mean Green function and $\Delta \Sigma$ is the fluctuation contribution to the self-energy part, determined from the relation $\Sigma = \langle V \rangle + \Delta \Sigma$. Note that $G(V)$ is independent of Σ , i.e. equation (9) is satisfied for an arbitrary Σ . Choosing Σ in the site- and frequency-diagonal form $\Sigma_{ijnn'} = \Sigma_{ii} \delta_{ij} \delta_{nn'}$ and replacing the function $G(\Sigma)$ in the second factor of the right-hand side of equation (9) by its site-diagonal part with elements $g_{ij}(\Sigma) = G_{ii}(\Sigma) \delta_{ij}$, and also neglecting a small change in the energy of the electron due to scattering by thermal fluctuations (single-site quasi-static scattering approximation), instead of (9) we obtain

$$G(V) \simeq G(\Sigma) [1 - (\Delta V - \Delta \Sigma) g(\Sigma)]^{-1}. \quad (11)$$

In contrast to (9), here the function $G(V)$ depends on Σ . Determining Σ from the condition that the total free energy of the system is minimum, we obtain the coherent-potential theory equation

$$\Delta \Sigma = \langle \Delta V [1 - g(\Delta V - \Delta \Sigma)]^{-1} \rangle. \quad (12)$$

In the case of ferromagnets (and paramagnets), the self-energy part Σ and the mean single-site Green function $\langle G(V) \rangle_{ii} \simeq G_{ii}(\Sigma) = g$ are independent of the site index i and spin (σ) diagonal. According to (10),

$$g_\sigma(\varepsilon) = \int \frac{v(\varepsilon')}{\varepsilon - \sigma \langle V_z \rangle - \Delta \Sigma_\sigma(\varepsilon) - \varepsilon'} d\varepsilon' \quad (13)$$

where $\nu(\varepsilon)$ is the non-magnetic DOS (per atom, band, and spin). From equation (12), for the fluctuation contribution $\Delta\Sigma_\sigma$, in the second order of field fluctuations ΔV we obtain the formula [30]

$$\Delta\Sigma_\sigma(\varepsilon) = \frac{g_\sigma^s(\varepsilon)\langle\Delta V_z^2\rangle}{1 + 2\sigma\langle V_z\rangle g_\sigma^s(\varepsilon)} + 2g_\sigma^s(\varepsilon)\langle\Delta V_x^2\rangle \quad (14)$$

where $g_\sigma^s(\varepsilon)$ is determined by expression (13) at $\Delta\Sigma_\sigma(\varepsilon) = 0$ and

$$\langle\Delta V_\alpha^2\rangle = \frac{1}{N_a} \sum_{\mathbf{q}n} \langle|\Delta V_{\mathbf{q}n}^\alpha|^2\rangle \quad (15)$$

is the mean square of the fluctuation of the on-site exchange field ('fluctuation', for short). In (15), N_a is the number of atoms, $\alpha = x, z$; the \mathbf{q} summation is carried out over the wavevectors within the Brillouin zone and the n summation is carried out over the thermodynamic frequencies $\omega_n = 2\pi nT$.

Taking into account (8) and (6), for the temperature-dependent contribution to the fluctuation $\langle\Delta V_\alpha^2\rangle$ we obtain a simple analytic formula

$$\langle\Delta V_\alpha^2\rangle = \frac{1}{N_a} \sum_{\mathbf{q}} \frac{UT}{2N\lambda_{\mathbf{q}}^\alpha} \frac{2}{\pi} \arctan \frac{U\varphi_{\mathbf{q}}^\alpha \pi^2 T}{6N\lambda_{\mathbf{q}}^\alpha} \quad (16)$$

where N ($=5$) is the number of d bands per atom and spin,

$$\lambda_{\mathbf{q}}^\alpha = 1 - U\chi_{\mathbf{q}}^\alpha(0) \quad \varphi_{\mathbf{q}}^\alpha = \left. \frac{d \operatorname{Im} \chi_{\mathbf{q}}^\alpha(\varepsilon)}{d\varepsilon} \right|_{\varepsilon=0}.$$

Deriving formula (16), we discarded the temperature-independent term, assuming that the zero-point fluctuations are already taken into account in the initial DOS $\nu(\varepsilon)$ calculated by the density-functional method and in the effective interaction constant U . Besides, we used an expansion of the complex function $\chi_{\mathbf{q}}(\varepsilon)$ at small thermal energies: $\chi_{\mathbf{q}}(\varepsilon) = \chi_{\mathbf{q}}(0) + i\varphi_{\mathbf{q}}\varepsilon$.

The equations for the fluctuations $\langle\Delta V_\alpha^2\rangle$, for the mean exchange field

$$\langle V_z \rangle = -Us_z \quad s_z = (n_\uparrow - n_\downarrow)/2 \quad (17)$$

and chemical potential μ

$$n_e = n_\uparrow + n_\downarrow, \quad (18)$$

where

$$n_\sigma = \frac{1}{\pi} \int \operatorname{Im} g_\sigma(\varepsilon) f(\varepsilon) d\varepsilon \quad (19)$$

is the number of electrons with spin projection σ and n_e is the total number of electrons (per atom, band), form a system of four non-linear equations ($f(\varepsilon) = [\exp((\varepsilon - \mu)/T) + 1]^{-1}$ is the Fermi function). At $T = 0$, the fluctuations $\langle\Delta V_\alpha^2\rangle$ vanish, and this system turns into the mean-field-theory system of equations (17) and (18). This gives one an opportunity to find the effective constant U from a known magnetic moment $m(0) = g\mu_B s_z(0)$; after that, at $T \neq 0$, our initial system makes up a closed system with respect to the variables $\langle\Delta V_x^2\rangle$, $\langle\Delta V_z^2\rangle$, $\langle V_z \rangle$ and μ . This system is only slightly more complicated than the one obtained in the static local approximation (SLA) [30]. In fact, taking account of dynamics and spatial correlations leads to modification of the formula for the spin fluctuations $\langle\Delta V_\alpha^2\rangle$ only. For the most part, the simple computational scheme of the SLA remains unchanged.

Solving the system of dynamic non-local equations at a fixed temperature, we calculate the mean DOSs $\nu_\sigma(\varepsilon, T) = \pi^{-1} \operatorname{Im} g_\sigma(\varepsilon, T)$, the magnetization $m(T) = g\mu_B s_z(T)$, the mean local magnetic moment

$$m_L(T)/m(0) = [(\langle V_z \rangle^2(T) + \langle(\Delta \mathbf{V})^2\rangle)/\langle V_z \rangle^2(0)]^{1/2}, \quad (20)$$

Table 1. The ferromagnetic (T_C) and paramagnetic (Θ_C) Curie temperatures and the effective (m_{eff}) and mean local ($m_L(T_C)$) magnetic moments of Fe–Ni invar, calculated in two approximations of the spin-fluctuation theory.

	T_C/T_C^{exp}	$\Theta_C/T_C^{\text{exp}}$	$m_{\text{eff}}/m_{\text{eff}}^{\text{exp}}$	$m_L(T_C)/m_L(0)$
SLA	0.72	0.74	0.43	0.87
DNA	0.84	0.90	0.93	0.90

Table 2. Experimental values of fundamental magnetic characteristics of Fe₆₅Ni₃₅ invar.

m_0^{exp} (μ_B) [34]	T_C^{exp}/k_B (K) [34]	$m_{\text{eff}}^{\text{exp}}$ (μ_B) [35]
1.8	520	3.3

and the enhanced static uniform susceptibility $\tilde{\chi}_0(0) = \chi_0(0)/(1 - U\chi_0(0))$. Comparing the susceptibility $N\tilde{\chi}_0(0)$ in the paramagnetic region with the Curie–Weiss law $\chi = m_{\text{eff}}^2/(3(T - \Theta_C))$, we determine the value of the effective magnetic moment m_{eff} and the paramagnetic Curie temperature Θ_C .

Note that our model is written for the case of N degenerate bands. It is easily proved that, in this case, one obtains the same equations as in the case of a single energy band with the only difference that the temperature T is replaced by T/N . The details of the calculations of the magnetic properties by the DNA of the SFT have been described in our papers [26, 27, 31].

3. Results and discussion

The invar effect occurs at a concentration around Fe₆₅Ni₃₅. Since order–disorder should not play a crucial role (this can be seen from the example of Fe₇₂Pt₂₈ [3]), the ordered structure Fe₃Ni can represent the Fe–Ni invar. We take the initial non-magnetic DOS from *ab initio* calculations for Fe₃Ni [32] performed in the local-density approximation by the augmented-spherical-wave method [33]. The constant sp background was eliminated from the initial DOS per atom, so that the area under the curve was equal to 10 (the number of d states per atom). Then the DOS was slightly smoothed out by convolution with the Lorentz function of half-width $\Gamma = 0.005W$ ($W = 8.94$ eV is the bandwidth) and normalized to one d band of unit width: $\int_0^1 \nu(\varepsilon) d\varepsilon = 1$. With the help of smoothing we take into account the damping of one-electron states resulting from electron–electron correlations. The smoothed DOS of the d band $\nu(\varepsilon)$ used for calculation is represented in figure 1. The number of d electrons per atom $N_e = Nn_e = 2N \int_0^{\varepsilon_F} \nu(\varepsilon) d\varepsilon$ (ε_F is the Fermi energy) is equal to 7.84. The effective interaction constant $u = U/N$ determined from an experimental value of the magnetic moment per atom $m_0^{\text{exp}} = Nm(0) = 1.8 \mu_B$ [34] is 1.34 eV. Note that applying the DOS of Fe₃Ni for the Fe–Ni invar, we also pursued a purely methodical objective to investigate the magnetic properties of Fe–Ni invar in the same calculation scheme as we used earlier [26] for Fe and Ni, i.e. we performed the calculations for Fe, Ni and Fe–Ni invar as far as possible in the same manner. We emphasize that the ground state of Fe₃Ni in the band calculations [32] is ferromagnetic.

The results of the calculation of the basic magnetic characteristics of Fe–Ni invar are represented in table 1 and figures 2–4. All of the characteristics are represented in units of their experimental values given in table 2.

Let us start from the SLA, in which expression (16) for fluctuations is replaced by

$$\langle \Delta V_\alpha^2 \rangle_{\text{SL}} = \frac{UT}{2N\lambda_L^\alpha} \quad \lambda_L^\alpha = 1 - U\chi_L(0) \quad (21)$$

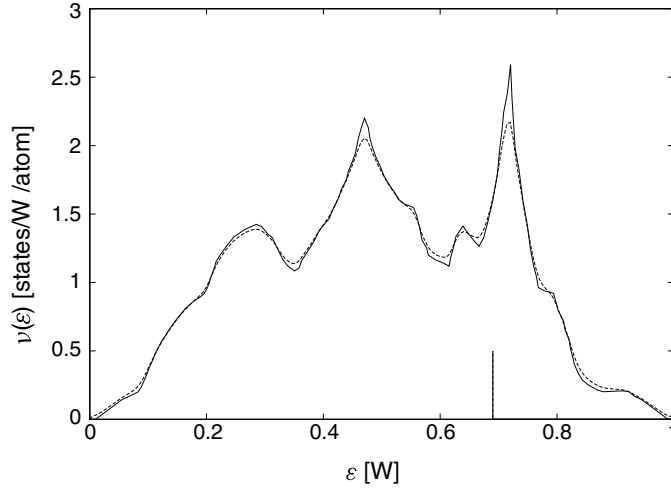


Figure 1. The DOS of the d band of non-magnetic Fe₃Ni, obtained from [32] (—), and the one smoothed out by convolution with the Lorentz function of half-width $\Gamma = 0.005$ (---). The energy ε and half-width Γ are in units of the bandwidth $W = 8.94$ eV. The vertical line indicates the position of the Fermi level ε_F .

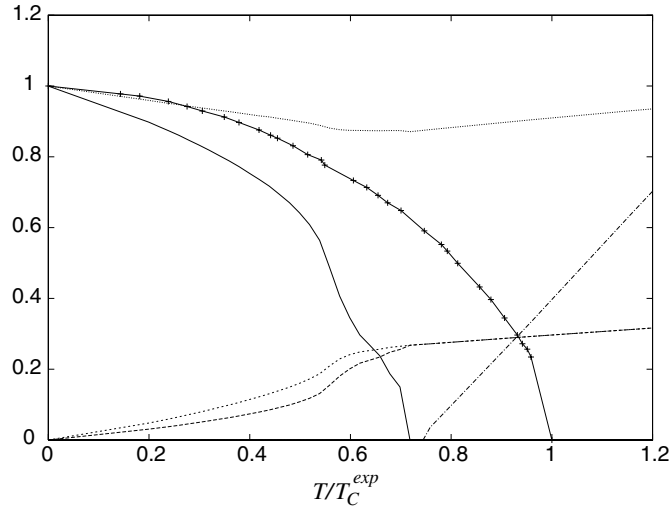


Figure 2. The magnetization $m(T)/m(0)$ (— calculation, +++ experiment [34]), the mean square of fluctuations of the on-site exchange field $\langle \Delta V_x^2 \rangle$ (- - -) and $\langle \Delta V_z^2 \rangle$ (- · -) in units of the mean square of the exchange field at $T = 0$, the inverse paramagnetic susceptibility $(N\tilde{\chi}_0(0))^{-1}$ (- · ·) in units of T_C^{exp}/μ_B^2 , and the mean local magnetic moment $m_L(T)/m_L(0)$ (·····) of Fe–Ni invar, calculated in the SLA as functions of the reduced temperature T/T_C^{exp} .

where $\chi_L(0) = N_a^{-1} \sum_{\mathbf{q}} \chi_{\mathbf{q}}(0)$ is the local susceptibility. As can be seen from figure 2, the magnetization $m(T)$ decreases too fast and the Curie temperature is much less than the experimentally observed one, $T_C = 0.72T_C^{\text{exp}}$. Moreover, a noticeable decrease of magnetization, $\propto T$, is seen at low temperatures, which is due to the fact that spin fluctuations increase linearly with temperature (see (21)). In general, the paramagnetic susceptibility

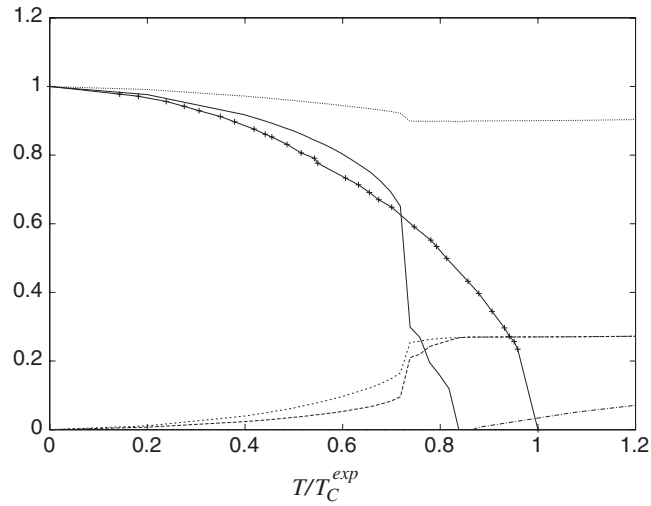


Figure 3. As figure 2, but calculated in the DNA.

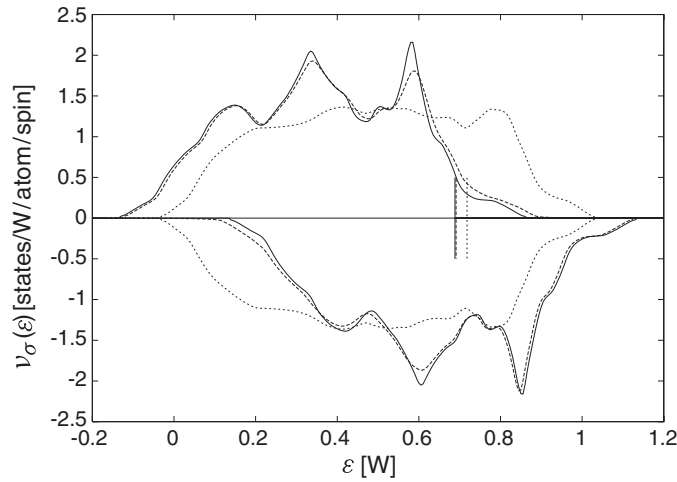


Figure 4. Spin-polarized DOSs of Fe–Ni invar at $T/T_C^{\text{exp}} = 0$ (—), 0.4 (---), and 0.84 (- - -), calculated in the DNA. The energy ε is given in units of the bandwidth W . The vertical line indicates the position of the chemical potential μ .

follows the Curie–Weiss law, but the effective magnetic moment m_{eff} is only 0.43 of its experimental value. The paramagnetic Curie point Θ_C , obtained by the linear extrapolation of $\tilde{\chi}^{-1}(T)$ to zero, is a little greater than the ferromagnetic one as it must be. Note that the linear decrease of the magnetization at low temperatures and a small effective moment in the paramagnetic region are common shortcomings of the calculations performed in the static approximation (see, e.g. [26] and references therein).

In the DNA the fluctuations $\langle \Delta V_\alpha^2 \rangle_{\text{DN}}$, calculated using formula (16), at low temperatures increase slowly ($\propto T^2$) with increasing temperature. For this reason, at low temperatures the magnetization is proportional to T^2 , which provides a good agreement with the experimental curve in the initial region (figure 3). The Curie temperature T_C obtained is equal to $0.84T_C^{\text{exp}}$.

The paramagnetic susceptibility follows the Curie–Weiss law and the effective magnetic moment is a little less than the experimental one: $m_{\text{eff}} = 0.93m_{\text{eff}}^{\text{exp}}$. The paramagnetic Curie point Θ_C is a little greater than T_C .

As can be seen from figure 3, the local magnetic moment $m_L(T)/m_L(0)$ calculated by formula (20) depends not too strongly on the temperature: with temperature increasing from zero to T_C^{exp} the local moment decreases by 10% only. However, in comparison with Fe, where the local moment on the same temperature interval remains almost constant [26], this change is considerable and quite sufficient for an explanation of the invar effect. The assumption that the volume change of the Fe–Ni invars is connected with the temperature variation of the local moment, and not the magnetization, was first made in [36] from an analysis of the experimental data. The subsequent SFT calculations [21, 22] confirmed this assumption.

Note that in the SFT for *weakly* ferromagnetic metals the decrease of the local moment at T_C reaches $\sim 23\%$ ($\eta(T_C) = m_L^2(T_C)/m_L^2(0) = 0.6$) [37]. Using $\eta(T_C) \sim 0.5$ and roughly estimating $D_0/B \simeq 10^{-6} (\text{emu/g})^{-2}$ in approximate equation (5) of [37] for the magnetic volume change ω_m in the weakly ferromagnetic limit,

$$\omega_m(T) = \frac{D_0}{B} M_0^2 [\eta(T) - 1] \quad (22)$$

where D_0 is the magneto-volume coupling constant for $q = 0$, B is the bulk modulus and M_0 is the uniform magnetization per atom at $T = 0$ K, for $\text{Fe}_{65}\text{Ni}_{35}$ the authors of [37] got $\omega_m(300 \text{ K}) \simeq -0.008$ in good agreement with the value deduced from experiment. However, from the preliminary calculation [37] it is not evident at all that in Fe–Ni invar such a large decrease of the local moment does occur. The fact is that the value of ω_m strongly depends on details of the calculation. So, using our $\eta = (0.9)^2$ and $D_0/B \simeq (2-3) \times 10^{-6} (\text{emu/g})^{-2}$ obtained in [37] by the theoretical estimation of the D_0 , for $\text{Fe}_{65}\text{Ni}_{35}$, which has $M_0 \simeq 170 \text{ emu g}^{-1}$ [3], from the same equation (22) we obtain $\omega_m(T_C) = -0.011$ to -0.016 also in good agreement with the results of the measurements [38]. In more exact SFT calculation [21] for $\text{Fe}_{0.6}\text{Ni}_{0.4}$, $\omega_m(T_C) = -0.06$ is obtained, which is too large compared with the experimental value -0.0136 [38]. Since in our calculation the local moment decreases noticeably more slowly than that in [21], for ω_m in the DNA of the SFT one can expect a better agreement with experiment. Our small decrease of the local moment at the Curie temperature is also supported by the ratio of the experimental values $\omega_m(T_C)$ for $\text{Fe}_{65}\text{Ni}_{35}$ and Fe: -0.0190 and -0.0014 , respectively, [39]. Finally, the assumption that in the single-site approximation [19, 20] the decrease of the local moment and hence the value of the magnetic volume change is overestimated was stated in [21, 40], where direct calculations $\omega_m(T_C)$ for Fe gave values -0.04 [21] and -0.03 [40], which is noticeably less than is obtained within the Stoner theory, but still much more than is observed experimentally. In [39] it is claimed that the reduction of the local moment of Fe at T_C must be only 1% in order to obtain a reasonable value of ω_m .

Since the local moment is the most important physical parameter in the invar problem, we emphasize that our 10% decrease is comparable to the 7% decrease obtained in SFT calculations [41]. Unfortunately there are no reliable *experimental* data on the temperature variation of the local moment. However, making a reasonable assumption that the local moment is a linear function of the atomic volume, from [42, 43] we see that the atomic volume change is significantly smaller (2%–3%) than was predicted in the commonly cited paper [8]. The authors of [8] find that a magnetization collapse is accompanied by a reduction of the atomic volume of $\sim 9\%$. (Their calculated lattice constant contraction is 3% as compared to the 0.7%–1% experimentally obtained in [42, 43].)

Figure 4 shows the mean DOSs $\nu_\sigma(\varepsilon, T)$ at zero, half Curie ($T = 0.4T_C^{\text{exp}}$) and Curie ($T_C = 0.84T_C^{\text{exp}}$) temperatures. In contrast to the mean-field-theory results, as the

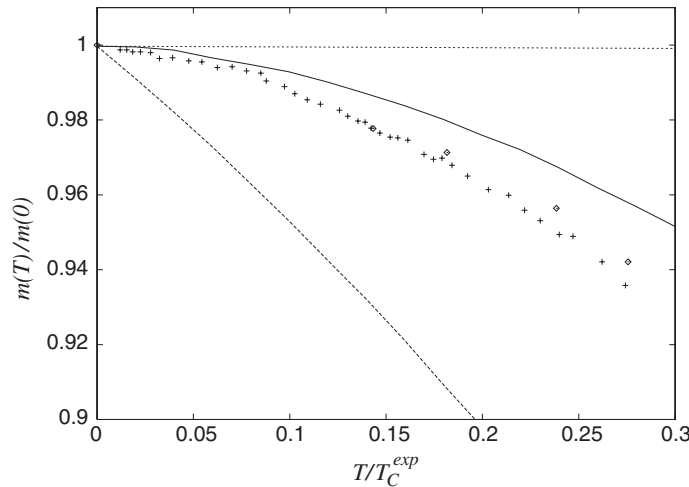


Figure 5. The magnetization of Fe–Ni invar at low temperatures, calculated in the mean-field approximation (---), SLA (— · —) and DNA (—). The crosses represent the experimental data [44] and diamonds represent the experimental data [34].

temperature increases, the functions $\nu_{\uparrow}(\varepsilon, T)$ and $\nu_{\downarrow}(\varepsilon, T)$, shifting toward each other, become noticeably smoothed.

Now we dwell on the temperature dependence of magnetization $m(T)$ at low temperatures, which is still an unsolved problem in the invar theory. The observed $m(T)$ with increasing temperature decreases much faster than would be expected from magnon excitations [44]. Some authors [44] explain the additional decrease of magnetization by unknown hidden excitations, others [45] by Stoner-type excitations. For explanation of the temperature behaviour of $m(T)$ in the framework of recent theory, we calculate in detail the magnetization of the Fe–Ni invar at low temperatures in the mean-field approximation ($\langle \Delta V_x^2 \rangle_{\text{MF}} = \langle \Delta V_z^2 \rangle_{\text{MF}} = 0$) and in two approximations of the SFT. As can be seen from figure 5, at low temperatures the contribution of Stoner excitations in the decreasing of $m(T)$ is negligible. In the SLA the magnetization decreases too fast ($\propto T$). Only in the DNA, in the case where the spin fluctuations are proportional to T^2 , a good agreement with the experimental data is obtained for the calculated $m(T)$.

4. Conclusion

Our numerical calculations of the magnetic properties of Fe–Ni invar demonstrate that the static approximation of the SFT overestimates the contribution due to spin fluctuations and yields an incorrect temperature dependence of the magnetization and susceptibility. Only simultaneous consideration of the dynamics and non-locality of the spin fluctuations ensures agreement of the theory with experiment.

Note that a success of our spin-fluctuation approach in the region of *intermediate* temperatures, where the single-site approximation is most appropriate, is not surprising. What is surprising is a good agreement of our calculations with experiment at *low* temperatures, where one would think the spin-wave approach (SWA) is preferable. Recall that in our model the short-range magnetic order is taken into account in the derivation of formula (16) for spin fluctuations only. This shortcoming seems to be not so substantial as the neglect of the longitudinal fluctuations in the SWA.

Finally, though the use of the ordered Fe₃Ni DOS as the initial one for the purposes of the first dynamic non-local calculation is justified, in the future we intend to investigate the effect of randomness in the spatial arrangement of Fe and Ni atoms [41, 46–48] on the magnetic properties of the Fe–Ni invar at finite temperatures.

Acknowledgments

The author is grateful to V I Grebennikov for useful discussions and to the referees of the paper for constructive criticism.

References

- [1] Wassermann E F 1990 *Ferromagnetic Materials* vol 5, ed K H J Buschow and E P Wohlfarth (Amsterdam: Elsevier) p 237
- [2] Wassermann E F 1991 *J. Magn. Magn. Mater.* **100** 346
- [3] Shiga M 1994 *Materials Science and Technology* part II, vol 3B, ed R W Cahn, P Haasen and E J Kramer (Weinheim: VHC) p 159
- [4] Wassermann E F 1997 *The Invar Effect: a Centennial Symposium* ed J Wittenauer (Warrendale, PA: Minerals, Metals and Materials Society) p 51
- [5] Weiss R J 1963 *Proc. R. Soc.* **82** 281
- [6] Moruzzi V L 1990 *Phys. Rev. B* **41** 6939
- [7] Entel P, Hoffmann E, Mohn P, Schwarz K and Moruzzi V L 1993 *Phys. Rev. B* **47** 8706
- [8] Abrikosov I A, Eriksson O, Söderlind P, Skriver H L and Johansson B 1995 *Phys. Rev. B* **51** 1058
- [9] Rueff J P, Shukla A, Kaprolat A, Krisch M, Lorenzen M, Sette F and Verbeni R 2001 *Phys. Rev. B* **63** 132409
- [10] James P, Eriksson O, Johansson B and Abrikosov I A 1999 *Phys. Rev. B* **59** 419
- [11] van Schilfgaarde M, Abrikosov I A and Johansson B 1999 *Nature* **400** 46
- [12] Brown P J, Neumann K-U and Ziebeck K R A 2001 *J. Phys.: Condens. Matter* **13** 1563
- [13] Moriya T 1985 *Spin Fluctuations in Itinerant Electron Magnetism* (Berlin: Springer)
- [14] Capellmann H (ed) 1987 *Metallic Magnetism* (Berlin: Springer)
- [15] Murata K K and Doniach S 1972 *Phys. Rev. Lett.* **29** 285
- [16] Wagner D 1989 *J. Phys.: Condens. Matter* **1** 4635
- [17] Mohn P, Schwarz K and Wagner D 1991 *Phys. Rev. B* **43** 3318
- [18] Kaul S N 1999 *J. Phys.: Condens. Matter* **11** 7597
- [19] Hubbard J 1979 *Phys. Rev. B* **19** 2626
Hubbard J 1979 *Phys. Rev. B* **20** 4584
- [20] Hasegawa H 1979 *J. Phys. Soc. Japan* **46** 1504
Hasegawa H 1980 *J. Phys. Soc. Japan* **49** 178
- [21] Hasegawa H 1981 *J. Phys. C: Solid State Phys.* **14** 2793
- [22] Kakehashi Y 1981 *J. Phys. Soc. Japan* **50** 2236
- [23] Stratonovich R L 1957 *Dokl. Akad. Nauk SSSR* **115** 1097
Stratonovich R L 1958 *Sov. Phys.—Dokl.* **2** 416 (Engl. Transl.)
- [24] Hubbard J 1959 *Phys. Rev. Lett.* **3** 77
- [25] Reser B I and Grebennikov V I 1998 *Phys. Met. Metallogr.* **85** 20
- [26] Reser B I 1999 *J. Phys.: Condens. Matter* **11** 4871
- [27] Reser B I 2000 *J. Phys.: Condens. Matter* **12** 9323
- [28] Reser B I 2002 *J. Phys.: Condens. Matter* **14** 1285
Reser B I 2003 *J. Magn. Magn. Mater. C* **258/259** 51
- [29] Hertz J A and Klenin M A 1974 *Phys. Rev. B* **10** 1084
- [30] Reser B I and Grebennikov V I 1997 *Phys. Met. Metallogr.* **83** 127
- [31] Reser B I 1996 *J. Phys.: Condens. Matter* **8** 3151
- [32] Williams A R, Moruzzi V L, Gelatt C D Jr and Kübler J 1983 *J. Magn. Magn. Mater.* **31–34** 88
- [33] Williams A K, Kübler J and Gelatt C D Jr 1979 *Phys. Rev. B* **19** 6094
- [34] Crangle J and Hallam G C 1963 *Proc. R. Soc. A* **272** 119
- [35] Matsui M, Adachi K and Chikazumi S 1980 *J. Appl. Phys.* **51** 6319
- [36] Shiga M and Nakamura Y 1969 *J. Phys. Soc. Japan* **26** 24
- [37] Moriya T and Usami K 1980 *Solid State Commun.* **34** 95

- [38] Hayase M, Shiga M and Nakamura Y 1973 *J. Phys. Soc. Japan* **34** 925
- [39] Shiga M 1981 *Physics of Transition Metals 1980 (Inst. Phys. Conf. Ser. vol 55)* p 241
- [40] Grebennikov V I and Turov E A 1986 *Dynamic and Kinetic Properties of Magnets* ed S V Vonsovskii and E A Turov (Moscow: Nauka) p 223 (in Russian)
- [41] Kakehashi Y 1988 *Phys. Rev. B* **38** 474
- [42] Schumann F O, Wu S Z, Mankey G J and Willis R F 1997 *Phys. Rev. B* **56** 2668
- [43] Schumann F O, Willis R F, Goodman K G and Tobin J G 1997 *Phys. Rev. Lett.* **79** 5166
- [44] Ishikawa Y, Onodera S and Tajima K 1979 *J. Magn. Magn. Mater.* **10** 183
- [45] Nakai I, Ono F and Yamada O 1983 *J. Phys. Soc. Japan* **52** 1791
- [46] Hasegawa H and Kanamori J 1971 *J. Phys. Soc. Japan* **31** 382
- [47] Schröter M, Ebert H, Akai H, Entel P, Hoffmann E and Reddy G G 1995 *Phys. Rev. B* **52** 188
- [48] Wang Y, Stocks G M, Nicholson D M C, Shelton W A, Antropov V P and Harmon B N 1997 *J. Appl. Phys.* **81** 3873