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LETTER TO THE EDITOR

Configurational interactions in disordered and ordered alloys: general relations and phenomenological analysis for the FCC-based Ni–Fe and Ni–Al systems

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Abstract. We discuss microscopic expressions for the configurational potentials V_n in alloys using the KKR formalism and cluster-cumulant expansions. We note that V_n must depend not only on the concentrations but also on the long-range order parameters η_1 in the alloy. The analysis of the experimental data on the short-range order, phase diagrams and thermodynamics of the FCC-based Ni–Fe and Ni–Al systems appears to show that in the $Ni_{1-x}Fe_x$ alloys the dependence of V_n on both x and η_1 is weak, while in the $Ni_{1-x}Al_x$ ones the variations of V_n with x and (or) ordering are significant.

To describe statistical properties of alloys, such as their ordering, decay, short-range order (SRO) effects etc, the use of Ising-like Hamiltonians is generally accepted, these being of the form (see e.g. [1–5])

$$H = NV_0 + \sum_{i\alpha} V_i^\alpha N_\alpha + \sum_{\substack{i < j \\ \alpha\beta}} V_2^{i\alpha,j\beta} n_{i\alpha} n_{j\beta} + \sum_{\substack{i < j < k \\ \alpha\beta\gamma}} V_3^{i\alpha,j\beta,k\gamma} n_{i\alpha} n_{j\beta} n_{k\gamma} + \dots \quad (1)$$

Here N_α is the number of atoms of the α species, $N = \sum_\alpha N_\alpha$ is the total number of atoms, the operator $n_{i\alpha}$ is unity when the site i is occupied by an α -species atom and $n_{i\alpha} = 0$ otherwise, and the coefficients $V_n^{i\alpha,\dots,j\beta}$ will be called the configurational potentials. In the substitutional alloys to be discussed below the operators $n_{i\alpha}$ at each i are bounded by the condition $\sum_\alpha n_{i\alpha} = 1$, thus in the binary alloy A–B the Hamiltonian (1) can be expressed, for example, in terms of the n_{iA} operators only.

In the original phenomenological approaches the potentials V_n in (1) were supposed to be constants independent of both the concentrations $c_\alpha = N_\alpha/N$ and the state of order in the alloy [1]. However, the microscopic estimates based on the coherent potential approximation (CPA) for the electronic structure of disordered alloys showed that the V_n values can vary significantly with the concentration [2–4]. The analysis of the SRO data for the alloys Ni_xCu_{1-x} [6], as well as the calculations of [7], confirmed these estimates, revealing sharp variations of V_n with x in these alloys (which is connected with the fact that the Fermi level ϵ_F approaches the peak in the density of electronic states, $N(\epsilon)$, when x increases). However, the possible changes of V_n under the ordering of the alloy were apparently not discussed in the literature. In particular, in estimates of the

energy differences between different phases [4, 5] the V_n values in the ordered and disordered phases were always supposed to be the same.

In this letter we note firstly that the potentials V_n in (1) must, generally, vary with ordering, and the variations can be not small if the corresponding changes of electronic structure are significant. Secondly, we present estimates of V_n from the experimental data for the FCC-based Ni-Fe and Ni-Al alloys, which, as they seem to be interesting themselves, may also illustrate the above considerations.

In the theoretical discussion we consider only the 'electronic' contribution to V^0 (that connected with the local lattice distortions is usually believed to be small for the substitutional alloys) and use the conventional approximations of the density functional theory (DFT) and the muffin-tin (MT) form for the electronic crystalline potential. Then the expression for the thermodynamic potential of electrons, $\Omega(\mu) = E - \mu N_e$, corresponding to their ground state at the given set $\{n_{i\alpha}\}$ of the occupation numbers in the KKR formalism has the form [8, 4]

$$\Omega - \Omega_0(\mu) = \frac{1}{N\pi} \text{Im Tr} \int_{-\infty}^{\mu} d\varepsilon \ln \tau + U^{\text{dc}}\{\rho_{i\alpha}\} + \sum_{i < j, \alpha\beta} \frac{Z_{\alpha} Z_{\beta}}{R_{ij}} n_{i\alpha} n_{j\beta} \quad (2a)$$

$$\tau = \left(\sum_{i\alpha} m_{i\alpha}^i n_{i\alpha} - \mathbf{g} \right)^{-1}. \quad (2b)$$

Here $\Omega_0(\mu)$ is the free-electron value of Ω ; $m_{i\alpha}^i = (t_{i\alpha}^i)^{-1}$ is the inverse of the scattering matrix at the site i occupied by the atom of species α ; $(t_{i\alpha}^i)_{ij}^{L'L'} = \delta_{ij} t_{i\alpha}^{L'L'}(\varepsilon)$, where L denotes the angular momentum variables; and \mathbf{g} is the KKR structural constant matrix. The term U^{dc} describes the 'double counted' terms of the electron-electron interaction; it is a functional of the electronic density in the MT cells, $\rho_{i\alpha}(r)$, which is related to the operator τ (2b) as [8]

$$\rho_{i\alpha}(r) = -\frac{1}{\pi} \text{Im} \int_{-\infty}^{\mu} d\varepsilon \sum_{LL'} Z_{i\alpha}^{L\alpha}(r) Z_{i\alpha}^{L'\alpha}(r) \tau_{LL'}^{i\alpha, i\alpha} \quad (3)$$

where $Z_{i\alpha}^{L\alpha}(r)$ is the wave function in the MT potential $V_{i\alpha}(r)$ [8]. The last term in (2a) corresponds to the Coulomb repulsion of ions, $R_{ij} = |\mathbf{R}_i - \mathbf{R}_j|$ being the intersite distance.

The expression (2a) can serve as a basis for the derivation of the configurational Hamiltonian (1) [2-4]; we can obtain microscopic expressions for $V_n^{\alpha\alpha\cdots\beta\beta}$, equating coefficients at $n_{i\alpha} \dots n_{j\beta}$ in equations (1) and (2a). For a system having a finite number of sites, such a definition would be unique. However, in the macroscopic system the expansion (1) is necessarily truncated after several terms with the smallest values of n and R_{ij} . Then the adequacy of using such a truncated Hamiltonian H_t to calculate the statistical properties is determined by the extent to which the results of the operation of H_t and the total H (1) on the configurations $\{n_{i\alpha}\}$, making the main contribution to the statistical sum $Z = \sum \exp(-H/T)$, are similar. In practice, expressions for H_t are obtained with the use of various versions of the 'effective medium' approximation (CPA, the embedded cluster method (ECM) [3, 6], the cluster field approximation [9], etc). They correspond to regrouping terms and partial averaging in equation (2a) aiming to reduce it to the form (1) with the coefficients $V_n^{\alpha\alpha\cdots\beta\beta}$ decreasing as rapidly as possible with an increase of n or R_{ij} . Hence, the V_n values must be determined self-consistently, depending, therefore, on both the concentrations and on the state of order of the alloy.

The cluster-cumulant expansions [10], analogous to those used in the statistical theory of alloys [11, 12], may provide a natural basis for the construction of various

approximations for calculating V_n . They correspond to introducing variational parameters into equations (2) which describe the effect of the surroundings on the n -site cluster under consideration. We shall consider them, using as an example the 'single-site' approximations, such as the CPA or ECM (though the many-site approaches are also possible [9]).

In the disordered alloy, each m_α^i in (2) is written as a sum of the site-independent term m_c and the deviation from it, $\Delta_\alpha^i = m_\alpha^i - m_c$:

$$m_\alpha^i = m_c + \Delta_\alpha^i \quad m_c = m_c^L(\varepsilon)\delta_{ij}\delta_{LL'}. \quad (4)$$

Then we write $\ln \tau$ in (2a) as a cumulant expansion:

$$\begin{aligned} \ln \tau = \ln \tau_0 + \sum_{i\alpha} n_{i\alpha} \ln Q_\alpha^i + \sum_{i < j, \alpha\beta} n_{i\alpha} n_{j\beta} (\ln Q_{\alpha\beta}^{ij} - \ln Q_\alpha^i - \ln Q_\beta^j) \\ + \sum_{i < j < k, \alpha\beta\gamma} n_{i\alpha} n_{j\beta} n_{k\gamma} (\ln Q_{\alpha\beta\gamma}^{ijk} - \ln Q_{\alpha\beta}^{ij} - \ln Q_{\beta\gamma}^{jk} - \ln Q_{\gamma\alpha}^{ki} \\ + \ln Q_\alpha^i + \ln Q_\beta^j + \ln Q_\gamma^k) + \dots \end{aligned} \quad (5)$$

where $\tau_c = (m_c - \mathbf{g})^{-1}$ and $Q_{\alpha\beta\dots\gamma}^{i\dots k} = [1 + (\Delta_\alpha^i + \Delta_\beta^j + \dots + \Delta_\gamma^k)\tau_c]^{-1}$. The analogous expansion can also be written for τ in (2b):

$$\tau = \tau_c \left[1 + \sum_{i\alpha} n_{i\alpha} (Q_\alpha^i - 1) + \sum_{i < j, \alpha\beta} n_{i\alpha} n_{j\beta} (Q_{\alpha\beta}^{ij} - Q_\alpha^i - Q_\beta^j + 1) + \dots \right]. \quad (6)$$

Inserting equations (5) and (6) in equation (2a) yields expansion (1). Hence, the term with the product of n operators $n_{i\alpha} \dots n_{j\beta}$ corresponds to the contribution to H of the electron scattering at n different sites in the effective medium. If the operator τ_c describes the 'averaged' motion of electrons adequately, one may expect that these n -site contributions decrease sufficiently rapidly with the increase of n or R_{ij} , so the convergence of expansion (1) for the significant configurations $\{n_{ij}\}$ is acceptable.

If all the terms of series (5) and (6) are taken into account, the Ω value in (2) does not depend on m_c , thus $\delta\Omega/\delta m_c = 0$. However, such a dependence is present for the 'truncated' $\Omega = \Omega_t$, corresponding to a finite number of terms in the expansion. One may expect that the error is minimal if we choose m_c to obey the condition $\delta\Omega_t/\delta m_c = 0$; this corresponds to variational formulations of the single-site approximations [13]. In varying the expression (2a) over m_c , the values of $\rho(\mathbf{r})$ and $m_\alpha^i(\rho)$ can be kept constant† since in the DFT used the functional $\Omega(\rho)$ obeys the variational equation $\delta\Omega/\delta\rho(\mathbf{r}) = 0$. Thus we can only vary τ_c and Δ_α^i over $m_c(\varepsilon)$ in the first, 'band' term of (5). Then allowing for only two first, single-site terms in equations (5) and (6) yields the CPA equations for m_c :

$$\sum_{i\alpha} n_{i\alpha} \Delta_\alpha^i (1 + \tau_c^0 \Delta_\alpha^i)^{-1} = N \sum_{\alpha} c_\alpha \Delta_\alpha (1 + \tau_c^{00} \Delta_\alpha)^{-1} = 0. \quad (7)$$

Here we took into account the 'self-averaging' of the sum over all the sites i in equation (7) and also the independence of m_α^i and Δ_α^i on the site number i used in the single-site approximation.

To find the configurational potentials V_n we must take into account the n -site cumulants with $n \geq 2$ in equations (5) and (6). Hence, m_c must be determined, generally

† This point was indicated to us by N E Zein.

speaking, from the same variational equation $\delta\Omega/\delta m_c = 0$, including these n -site contributions to Ω , while in the previous calculations of V_n [2-4, 7] the m_c values were found from the CPA single-site equation (7). However, if the many-site terms are supposed to be smaller than the single-site ones by some parameter δ , then the contribution to Ω from the correction $m_c - (m_c)_{\text{CPA}}$ has the order δ^2 and neglecting it is justified.

In the ordered alloy the lattice sites i are divided into several non-equivalent sublattices i_λ with the different concentrations $\langle n_\alpha(i_\lambda) \rangle = c_\alpha^\lambda$. Thus, to adequately describe the electronic structure, the single-site CPA parameters $m_c(i_\lambda) = m_c^\lambda$ for different sublattices must be different [14] and can be found from the set of equations $\delta\Omega/\delta m_c^\lambda = 0$. Therefore, the averaged quantities $\langle \Delta_c^i \rangle = \Delta_c^\lambda$ and $\tau_c^{ii} = \tau_c^{00,\lambda}$ in the equations generalizing (7) depend on the type λ of the sub-lattice. It is clear from equations (2)-(7) that the potentials $V_n^{i_\lambda \alpha \dots j_\mu \beta} = V_n(i_\lambda \alpha, \dots, j_\mu \beta)$ in (1) for different sublattices λ, \dots, μ are different too. In addition, all these potentials, including the 'configuration-independent' terms V_0 and V_α in (1), depend on all the c_α^λ and in particular, on the order parameters η_i in the alloy.

The previous treatments [2-5] did not consider the changes of V_n with ordering, supposing $V_n(\eta_i) = V(0)$. This is evidently reasonable if the changes of the electronic structure under the ordering are small (for example, when the constituents of the alloy have similar band structures). If, however, these changes are significant (as, for example, in the FeTi alloy [14]) then the V_n values must, generally, change noticeably under the ordering.

Experimentally, the V_n values can be estimated (in that or another approximation) from the phase diagram and thermodynamic data [15, 16] or, more directly, from the SRO studies [6, 11]. Below we present such estimates for the FCC-based Ni-Fe and Ni-Al alloys. Hence, to calculate statistical properties using the Hamiltonian (1), we used the cluster field method (CFM) described earlier [11, 12]. This is a simplified version of the known cluster variation method (CVM) [1] which is also convenient for considering the long-range interactions. Detailed analysis [11, 12] has shown that for the V_n values characteristic of the considered alloys, the accuracy of the employed versions of the CFM is rather high and, apparently, exceeds that of the experimental data used.

Estimates of V_n in the FCC alloys $\text{Ni}_{1-x}\text{Fe}_x$ were discussed in detail in papers [6] and [12]; here we summarize the results. The SRO was studied in the disordered A1 phase with the diffuse scattering of neutrons at $x = 0.235$ [17] and $x = 0.698$ [18]. Since only pair correlation parameters are determined with this method, one must make some assumptions about the non-pair, many-site terms in (1). The assumption of pure pairwise interactions, $V_{n \geq 3} = 0$, results in very sharp concentration dependencies of the potentials V_2^{ij} (see table 1) and in disagreement with the observed phase diagram (x, T) in the region $x \approx 0.25$ [6, 12]. At the same time, the presence of the three-site interaction of nearest neighbours $V_3^{ijn} = (200-250)$ K leads to much less variation of V_2^{ij} between $x = 0.235$ and $x = 0.698$. Figure 1 shows the phase diagrams calculated with these V_3 s assuming both V_3 and V_2^{ij} to be concentration independent. The satisfactory agreement with experiment, seen in this figure, can be considered as a substantiation of the assumptions about weak variations of the V_n in $\text{Ni}_{1-x}\text{Fe}_x$ with both x and orderings.

Note also that the ordering of the alloy results in the appearance of additional gaps or pseudogaps in the electron energy spectrum which usually correspond to some gain in the band energy. This gain, disregarded in the employed model, must enhance the transition temperatures $T_c(x)$, i.e. improve the agreement with experimental findings shown in figure 1. If one ascribes underestimating the calculated $T_c(x)$ in figure 1 to this

Table 1. Pairwise configurational potentials $V_{AA}^i + V_{BB}^i - 2V_{AB}^i = V_2(R_{ij})$ (in K) in the disordered FCC alloys $\text{Ni}_{1-x}\text{Fe}_x$ and $\text{Ni}_{1-x}\text{Al}_x$ as estimated from the SRO data [17, 18, 20] at various values of the three-site nearest-neighbour potential V_3^m .

Alloy	x	T (K)	V_3^m (K)	$2R_{ij}/a$									
				110	200	211	220	310	222	321	400	330	411
Ni-Fe [17]	0.235	780	0	663	-238	42	27	-22	-19	15	-9	8	-8
			-200	767	-229	44	29	-22	-19	15	-9	8	-8
			-250	797	-225	45	29	-22	-19	15	-9	8	-8
Ni-Fe [18]	0.698	743	0	65	-146	-18	0	-7	-14	—	—	—	—
			-200	651	-145	-18	0	-7	-14	—	—	—	—
			-250	795	-147	-18	0	-7	-14	—	—	—	—
Ni-Al [19]	0.073	673	0	1188	-258	8	-186	-73	131	80	42	-11	-10
	0.105	673	0	1300	-249	-121	-251	-35	61	38	105	-21	10
		823	0	1341	-294	-109	-256	-12	48	41	116	-26	-34

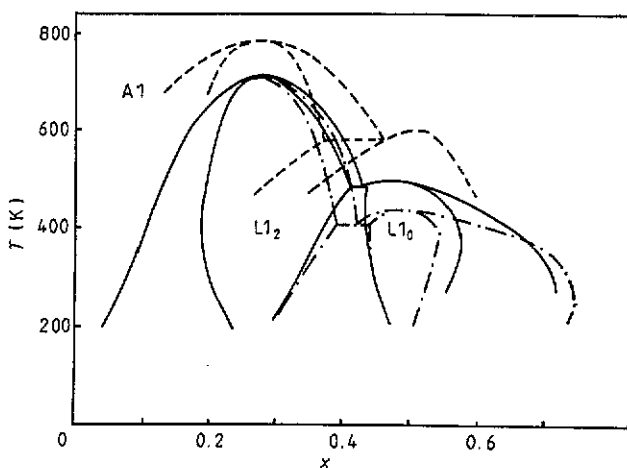


Figure 1. Phase diagram of the FCC alloys $\text{Ni}_{1-x}\text{Fe}_x$. Broken curve: results of Rossiter and Jago [29], full curve: calculated using V_n from the second line of table 1, chain curve: calculated using V_n from the third line of table 1.

effect only, then the changes of V_n in $\text{Ni}_{1-x}\text{Fe}_x$ under the $L1_2$ - and $L1_0$ -type orderings are estimated to be off by about 10–20%.

Therefore, the variations of V_n in the FCC alloys $\text{Ni}_{1-x}\text{Fe}_x$ with both x and orderings are apparently not large, which may reflect the similarity of the electronic structure of the constituents. The presence of noticeable three-site interactions can be connected with the itinerant magnetism of these alloys, i.e. with some non-pair collective effects.

In estimating V_n for the FCC Ni–Al system we used the so-called ‘5’ version of the CFM. It exceeds in accuracy the ‘6’ version described earlier [11] taking into consideration all the five-site clusters in which any two sites are connected with at least one chain of nearest neighbours. For the model of the A_3B alloy with pair interaction of nearest

neighbours only, $V_2^n = V$, the temperature of the L1₂-type ordering within the $\bar{5}$ approximation is $T_c = 0.41, V$, while the Monte Carlo calculations [19] yield: $T_c \approx 0.45 V$. When the ratio T/V grows, the accuracy of the CFM rapidly increases [11, 12], thus for the Ni–Al alloys, where $T_c/V > 1$, it must be sufficiently high.

Estimates of V_n from the SRO data [20] for the disordered γ phase of the Ni–Al alloys are presented in table 1. The potentials V_n were supposed to be pairwise (in accordance with the available estimates for non-magnetic alloys [2, 3]) and we assumed only ten V_2^j , with the smallest R_j values, to be non-zero. If we assume only five $V_2^j \neq 0$, then, for example, in the eighth line of table 1 we obtain $V_2^j = (1466, -221, -121, -235, -58)$ K, which is close to the results of the inverse Monte Carlo method [20] for the model with four $V_2^j \neq 0$: $V_2^j = (1439, -216, -137, -253, 0)$ K. This confirms the high accuracy of the $\bar{5}$ approximation in the problem under consideration.

The main interactions V_2^j for Ni_{1-x}Al_x in table 1 are almost constant with T ; this may indicate [6, 11] the adequate accuracy of the SRO data used [20], in spite of the difficulties in subtracting the background as discussed by the authors. Variations of V_n with x between $x = 0.073$ and $x = 0.105$ do not seem to be significant and may lie within experimental error.

In describing thermodynamics, we proceed from the expression for the free energy F (per atom of alloy) that neglects all the anharmonic effects [21]

$$F = F_{ci}(x) + F_{conf}(x, T) + F_{ph}(x, T). \quad (8)$$

Here the configuration-independent term F_{ci} corresponds to terms with V_0 and V_1 in (1), F_{conf} is the configurational contribution to F , and F_{ph} , the phonon contribution. To estimate the concentrational dependence of F_{ph} we employ the Debye model and data [22] on elastic constants C_{ij} in Ni_{1-x}Al_x, which reveal very small changes of C_{ij} between pure Ni ($x = 0$) and the L1₂-ordered γ' phase Ni₃Al ($x = 0.25$). If we assume the x dependence of the Debye temperature θ in this interval to be linear, then data [22] yield $\theta(x) = 459(1 + 0.072x)$ K. This corresponds to negligibly small variations of F_{ph} with x in both the γ and γ' phases.

The function $F_{ci}(x)$ was interpolated by a polynomial [16]

$$\Delta F_{ci}^{\gamma}(x) = F_{ci}^{\gamma}(x) - F_{ci}^{\gamma}(0) = \frac{1}{2}ax^2 + bx. \quad (9)$$

Values of a and b in the γ phase were estimated from the thermodynamical data [23] using $F_{conf}(x, T)$ calculated with V_n from the eighth line of table 1 (assuming that they do not change with x). This gave $a = 39173$ K and $b = -17483$ K. This estimate implies, in particular, that the x dependence of F is mainly determined by the F_{ci} term. For example, at $x = 0.19$, $T = 1633$ K we have $\mu_{ci} = \partial F_{ci}/\partial x = -9962$ K and $\mu_{conf} = \partial F_{conf}/\partial x = 944$ K.

If we now assume that the estimated V_n , a and b values in Ni_{1-x}Al_x at $x \leq 0.25$ do not change with either x or ordering, then the calculated phase diagram γ – γ' disagrees significantly with the observed one, see figure 2. This may correlate with the noticeable changes of electronic structure in these alloys: according to calculation [24], the form of $N(\epsilon)$ in the vicinity of the Fermi level in Ni₃Al differs significantly from that for pure Ni (see, e.g. [25]) which may result in considerable changes in V_n [6, 7].

To obtain an idea of the scale of these changes, we made illustrative estimates using only the phase diagram and long-range order data for the γ' phase [26–28]. Let us assume that the ΔF_{ci} term in this phase has the form not of equation (9), but

$$\Delta F_{ci}(x, \eta) = F_{ci}(x, \eta) - F(0, 0) = \frac{1}{2}Ax^2 + Bx + C + Dx\eta. \quad (10)$$

Here η is the order parameter related to the Al concentrations c_{Al}^{λ} in sublattices a and

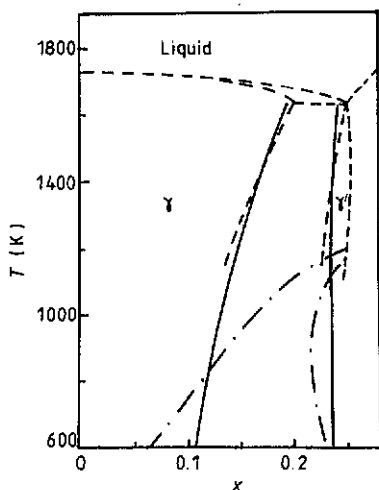


Figure 2. Phase diagram of the FCC alloys $\text{Ni}_{1-x}\text{Al}_x$. Broken curve: experimental results from [26] and [27], chain curve: calculated using ΔF_{ci} from equation (9) and V_n from the eighth line of table 1 for both the γ and γ' phases, full curve: calculated using ΔF_{ci} and V_n for the γ' phase changed as described in the text.

b , occupied predominantly by Ni and Al atoms, as $c_{\text{Al}}^a = x(1 - \eta)$ and $c_{\text{Al}}^b = x(1 + 3\eta)$. Equation (10) may be considered as an expansion of ΔF_{ci} in powers of the parameter $1 - \eta$ which is small in the γ' phase [28]. Furthermore, V_2^{ij} for the not-nearest neighbours are supposed to be the same as in eighth line of table 1, while those for the nearest neighbours were chosen to be $V^{ab} = 2500$ K and $V^{aa} = 1000$ K. Then the D value in (10) was fitted to the observed $\eta \approx 0.7$ at $x = 0.24$, $T = 1633$ K [28]; afterwards A , B and C were fitted to the phase diagram data [26, 27]. This gave (in K), $A = 71322$, $B = -29618$, $C = 1750$ and $D = -213$. Thus, the form of $\Delta F_{ci}(x, \eta)$ in the γ' phase differs noticeably from that in the γ phase.

The resulting phase diagram is shown in figure 2. Note that the choice of V^{ab} in the model used is restricted (V^{aa} is less important due to smallness of $1 - \eta$): at $V^{ab} \geq 3000$ the η value always exceeds the observed $\eta \approx 0.7$, while at $V^{ab} \leq 2000$ coefficients in equations (9) and (10) differ too strongly. For example, at $V^{ab} = 2000$ we obtain: $A \approx 113000$.

Let us comment on other estimates of V_n in $\text{Ni}_{1-x}\text{Al}_x$. Sigli and Sanchez considered only one potential, $V = 3890$ K, and the same ΔF_{ci} in both the γ and γ' phases, but their model does not describe the SRO data [20]. Cenedese *et al* [29] estimated V_2^{ij} from the data [20] using a combination of the CVM and Bragg-Williams method. However, they did not consider the thermodynamic data [23] and the ΔF_{ci} term; in addition, their calculated phase diagram disagrees with the observed one in a number of significant details.

Thus, the configurational potentials in the $\text{Ni}_{1-x}\text{Al}_x$ alloys appear to vary noticeably with x and/or the ordering, which may be connected with the aforementioned significant changes of the band structure. Since, however, this conclusion is based mainly on the analysis of the SRO data [20], the further SRO measurements for $\text{Ni}_{1-x}\text{Al}_x$, and in particular those at larger x , $x > 0.1$ and those obtained using the neutron scattering methods [6], seem to be highly desirable.

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