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LETTER TO THE EDITOR

A new method of calculating order–disorder transition temperatures and correlations in tight-binding alloys

T A Grishchenko, I V Masanskii and V I Tokar

Institute of Metal Physics, Academy of Sciences of the Ukrainian SSR, Vernadsky str. 36, 252680, Kiev-142, USSR

Abstract. We develop a simple method to calculate short-range order and temperatures of order–disorder transitions in binary alloys using a series expansion in powers of a parameter $\gamma = \exp(-1/\xi)$, where ξ is the dimensionless correlation length of the one-electron Green function. In the zeroth order our approach gives the theory of Ducastelle and Treglia (coherent-potential and Bragg–Williams approximations). In the lowest non-trivial order we obtain the spherical model results for transition temperatures and correlations in the disorder phase, with the effective nearest neighbour interaction potential identical to that of the generalised perturbation method. It turns out that in this order the corrections to the zeroth-order electronic Green function can be neglected, so we have the calculation scheme based on the well-known coherent-potential approximation.

Calculation of order–disorder transition temperatures and phase diagrams of binary alloys is a long-standing problem (for a review, see de Fontaine 1979). Usually the thermodynamic potential as a function of order parameters is computed. The configurational entropy is calculated within the cluster variation method (CVM) (Kikuchi 1951) while the internal energy is obtained both phenomenologically and by taking into account the electronic structure. The first approach is based on the Ising model, with the interatomic potentials regarded as input parameters (de Fontaine 1979). There exist several microscopic approaches, such as the Connolly and Williams (1983) technique, the generalised perturbation method (GPM) (Bieber *et al* 1981), the cluster-Bethe-lattice method (CBLM) (Robbins and Falicov 1982) and the KKR–CPA embedded-cluster method (Gonis *et al* 1984). However, the CBLM relies on severe topological approximation while in other methods an extrapolation from specific alloy configurations is used. In principle, we have to treat the electronic and atomic degrees of freedom on an equal footing and calculate the full partition function. Such a theory was developed by Ducastelle and Treglia (1980). Nevertheless, in their approach the short-range order (SRO), which plays an important role, is totally neglected. In this letter we propose a new method of calculating critical temperature which takes into account the presence of SRO and gives the above theory in the zeroth order.

    In our approach the series expansion in powers of a parameter $\gamma = \exp(-1/\xi)$ is used where ξ is a correlation length of the one-electron Green function measured in units of distance between nearest neighbour (NN) sites (Tokar 1985). This expansion is based on

the exponential decrease of the Green function with increasing distance between lattice sites i and j (see, for instance, Baxter 1982, p 26)

$$g_{ij} = F(i-j) \exp(-|i-j|/\xi) = O(\gamma^{|i-j|})$$

where $F(i)$ is a function varying at a rate less than exponentially.

We consider a tight-binding model Hamiltonian with diagonal disorder,

$$H = \sum_{ij} t_{ij} c_i^+ c_j + \sum_i (\varepsilon_i - \mu) c_i^+ c_i \tag{1}$$

where c_i is the annihilation operator of an electron at the site i, t_{ij} are the non-random transfer integrals, ε_i the random atomic levels taking the values ε^α, where $\alpha = \mathrm{A, B}$ and μ is the electronic chemical potential.

The corresponding partition function is (Vasil'ev 1976, p 16)

$$Z[a, a^+, A] = C \int D\psi D\psi^+ D\varphi \exp[S(\psi, \psi^+) + F(\varphi) + \psi^+ a + a^+ \psi + \varphi A] \tag{2}$$

where the action has the form

$$S(\psi, \psi^+) = \psi^+ (-\partial/\partial t - \mathcal{H})\psi \tag{3}$$

and

$$\exp F(\varphi) = [\delta(\varphi + \bar{\varepsilon} - \varepsilon^{\mathrm{A}}) + \delta(\varphi + \bar{\varepsilon} - \varepsilon^{\mathrm{B}})] \exp(\beta \nu \varphi).$$

ψ, ψ^+ and φ (a, a^+, and A) are fermion and boson (source) fields, the fermion ones depend on imaginary time, $\mathcal{H}$ is the corresponding one-particle Hamiltonian in the site representation, β the inverse temperature and ν the atomic chemical potential. To proceed with the irreducible correlation function we have introduced the average $\bar{\varepsilon}$ of the boson field and its fluctuation φ. In (2) and (3) reduced notations imply summation over site indices and integration over time from zero to β.

The partition function (2) may be written in the form (Tokar 1985)

$$Z[a, a^+, A] = C \exp(-a^+ g a) \exp(\tfrac{1}{2} A G A) R(ga, a^+ g, GA) \tag{4}$$

where

$$R(\psi, \psi^+, \varphi) = \exp[(\delta/\delta\psi) g (\delta/\delta\psi^+)] \exp[\tfrac{1}{2}(\delta/\delta\varphi) G (\delta/\delta\varphi)]$$
$$\times \exp[-\psi^+(\varphi + \bar{\varepsilon})\psi + \psi^+ \sigma \psi - \tfrac{1}{2}\varphi \Sigma \varphi + F(\varphi)]$$

is a generating functional for the S-matrix

$$g = (-\partial/\partial t - H - \sigma)^{-1} \qquad G = -\Sigma^{-1} \tag{5}$$

are exact fermion and boson Green functions, respectively, σ and Σ are corresponding self-energies. For fermion fields, left derivatives (Vasil'ev 1976, p 11) have been used.

From (4), similarly to Tokar (1985), we obtain the following equations for σ and Σ:

$$\delta^2 \ln R/\delta\psi\delta\psi^+ |_0 = 0$$

$$\delta^2 \ln R/\delta\varphi\delta\varphi |_0 = 0$$

$$\delta \ln R/\delta\varphi |_0 = 0$$

where subscript '0' denotes the condition $\psi = \psi^+ = \varphi = 0$.

Perturbation theory gives the following results: in the zeroth-order approximation we have the theory of Ducastelle and Treglia (1980); the coherent-potential approximation (CPA) equation

$$\sum_{\alpha} c^{\alpha} t^{\alpha} = 0 \qquad t^{\alpha} = (\varepsilon^{\alpha} - \sigma_0)/[1 - g_0(\varepsilon^{\alpha} - \sigma_0)]$$

(c^{α} is the concentration of α-atoms and t^{α} the scattering operator) and the Bragg–Williams one. Hereafter the subscripts '0' and '1' denote the diagonal and NN matrix elements, respectively. For the non-diagonal self-energies we obtain $\Sigma_1 = O(g_1^2)$, $\sigma_1 = O(g_1^3)$. The lowest-order expression for Σ_1 is

$$\Sigma_1 = \beta W_1/\Delta^2 \tag{6}$$

where

$$W_1 = (1/\pi) \int dE f(E) \, \mathrm{Im}[(\Delta t)^2 g_1^2]|_{E^+} \tag{7}$$

is the effective NN interaction potential (Bieber *et al* 1981) arising in the GPM as a result of elimination of electronic degrees of freedom, $f(E)$ is the Fermi function, $\Delta = \varepsilon^A - \varepsilon^B$ and $\Delta t = t^A - t^B$. From (6) and the link (5) between the boson Green function and the self-energy, the following expression for the Warren–Cowley SRO parameter α (Krivoglaz and Smirnov 1964) which is connected with G by

$$G = c(1 - c)\alpha\Delta^2$$

is obtained:

$$a_{ij} = P_{00}^{-1}(x)P_{ij}(x)$$

where x is found from the equation

$$P_{00}(x) = qc(1 - c)\beta W_1$$

and q is the coordination number and

$$P_{ij}(x) = \Omega^{-1} \int \frac{d^3k}{x - \lambda(k)} \exp[-i\,k(i - j)] \qquad \lambda(k) = q^{-1} \sum_{NN} \exp(i\,kj)$$

is the NN interaction Green function. Integration is over the Brillouin zone of the volume Ω. Note that in our approach the identity $\alpha_{00} = 1$ is automatically satisfied (for a discussion of this, see Ducastelle and Treglia 1980).

When the second-order transition with the wavevector k_0 takes place, we have at the critical point $\alpha^{-1}(k_0) = 0$ and, therefore, the transition temperature T_c is determined from (8) with $x = \lambda(k_0)$. These results correspond formally to the spherical model ones (Baxter 1982, p 74). However, the potential W_1 is expressed in terms of electronic structure parameters and depends on concentration and temperature.

Our preliminary numerical results are shown in figures 1 and 2. We have considered the equiatomic ordering alloy on the simple cubic lattice with a half-filled band and NN transfer. The Hamiltonian (1) is usually used for the description of d electrons. Therefore, we have taken into account the band (and spin) degeneracy and this leads to a factor of 10 entering (7). All energy values are measured in units of half-bandwidth. In figure 1, the NN SRO parameters given by

$$\alpha_1 = x - P_{00}^{-1}(x)$$

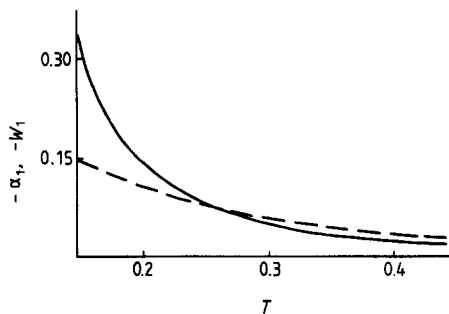and the potential W_1 are plotted versus temperature in the region from T_c to $3T_c$. The

Figure 1. Temperature dependence of the SRO parameter α_1 (full curve) and effective NN interaction potential W_1 (broken curve) in the region from T_c to $3T_c$ for $\Delta = 0.8$.
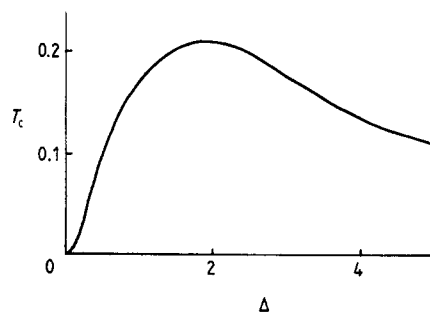

Figure 2. Critical temperature T_c as a function of the disorder parameter $\Delta = \varepsilon^A - \varepsilon^B$.

noticeable temperature dependence of the potential arises from a rapid change of the integrand in (7) near the Fermi level. Critical temperature as a function of the disorder parameter Δ is shown in figure 2. This curve has a qualitatively correct form (see, for example, Plischke and Mattis 1973). More detailed calculations are now in progress.

To summarise, we have obtained simple equations for calculating SRO parameters and order–disorder transition temperatures in binary tight-binding alloys. Note that in the lowest order the corrections to the zeroth-order electronic Green function can be neglected, and thereby we obtain a calculation scheme based on the well-known CPA.

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