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Correlation function approach to multicomponent systems: Three-body potentials in ternary alloys

G. Bruce Taggart*
Department of Physics, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

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With the use of the correlation function approach to multicomponent systems, the transition temperature is calculated within the mean-field approximation for a ternary alloy having arbitrary two-body and three-body interactions and arbitrary concentrations. This expression is studied in certain limiting cases. The concept of an effective concentration-dependent two-body interaction caused by the presence of three-body potentials is given justification. However, as has been pointed out for binary alloys and for mean-field approximations in general, this concept loses validity as the system approaches criticality and long-range correlations become dominant.

I. INTRODUCTION

The study of multicomponent solid solutions in general, and binary alloys in particular, has long occupied the experimental and theoretical interests of physicists and metallurgists.[1] While much work has been done on binary alloys, the literature on ternary alloys is sparse in comparison. Of late, however, interest in ternary alloys, as well as in other multicomponent systems, has increased due to a wealth of systems that have unusual or promising physical characteristics. As examples we might mention the high superconducting transition temperatures associated with the Chevrel phases,[2] the properties of Nb-W metal oxides,[3] the magnetic behavior of $Dy_{12}Fe_{32}O_2$,[4] the semiconducting properties of Ge-doped GaAs,[5] and such traditional studies as ordering in Au-Cu-Zn alloys.[6]

With this renewed interest in multicomponent systems has come the necessity of understanding on a microscopic level the origins of their physical properties. A first step in this direction would be to consider the effects of structure on static properties such as the transition temperature, the short- and long-range order parameters, and other equilibrium thermodynamic parameters. In particular we will look at the disordered ternary alloy as the simplest nontrivial multicomponent system, and determine the effects of irreducible static three-body potentials on the transition temperature.

Over the years a number of people have contributed to the theory of ternary alloys. Meijering[7] thoroughly studied segregation in ternary mixtures based on the free energy of a regular solution. Wojciechowski[8] seems to have been one of the first to attempt to extend the Bragg-Williams method from the binary alloy to the ternary alloy of arbitrary concentration, though a number of others considered the addition of a small amount of a third component to a binary mixture.[9] Murakami, Kachi, Nakanishi, and Takehara,[10] using the Bragg-Williams method, have derived self-consistent equations for the ternary alloy. With their result for the transition temperature Murakami *et al.* are able to reproduce a number of phase diagrams for three-component systems. The transition temperature calculated by Murakami *et al.*, which by the way reduces to Meijering's result in the regular solution limit, has also been obtained by de Fontaine[11] and Ryzhkov[12] using the more general method of free energy expansions in reciprocal space. Tahir-Kheli,[13] using Green's function techniques, has also obtained the result of Murakami *et al.*

All of the above theoretical results have been calculated in the ordered regime and, with the exception of de Fontaine and Ryzhkov, have *a priori* assumed the existence of sublattices. In the spirit of static concentration waves,[14] de Fontaine and Ryzhkov have worked in reciprocal space and have determined the transition temperature by looking for singularities in a free energy functional. Another approach to ternary alloys that also investigates the transition region in reciprocal space, but by means of the disordered regime, is that of Tahir-Kheli.[15] In this method correlation functions are calculated using high temperature series expansion techniques, and singularities in the spatial correlation function in reciprocal space determine the transition region. This technique has the advantage that one avoids calculating the system free energy and begins simply with the configurational energy. Misinterpretations[16] associated with this procedure have recently been removed,[17] with the result that critical properties calculated using this correlation function approach in the disordered regime agree with previous calculations in the ordered regime, e.g., the calculation of Murakami *et al.*

The ternary alloy calculations described above, whether in the ordered or disordered regime, have for the most part used the mean-field approximation, and have assumed that the interactions between atoms are central and pairwise (i.e., two-body potentials). However, it is well known for the binary alloy that theoretical methods that go beyond the mean-field approximation[1,18,19] significantly improve the description of real systems. Unfortunately such treatments of ternary alloys are rather limited in number.[20] Likewise the influence of interactions other than central pairwise interactions on the transition region of alloys has received even less attention in the literature. It is our intent here to consider specifically the influence of three-body interactions on the transition temperature of a ternary alloy. We leave for future considerations the influence of effects beyond the mean-field approximation.[21]

In Sec. II we shall consider the influence of three-body interactions in alloys. Following this in Sec. III the problem of a ternary alloy with static irreducible three-body potentials will be formulated, and for the disordered regime, the two-site correlation functions will be given in the mean-field approximation. In Sec. IV the correlation function approach to multicomponent alloys will be briefly described and in Sec. V the transition temperature will be given as a function of the static irreducible two-body and three-body interactions and arbitrary concentration. This result will be discussed in terms of appropriate limits and the concept of an effective concentration-dependent two-body interaction will be discussed. A brief summary will follow in Sec. VI.

II. THREE-BODY POTENTIALS

A comprehensive overview of the problem of many-body interactions in alloys has been given by Clapp.[22] We refer the reader to this review in which a number of justifications are given for the existence of many-body interactions in alloys. As a partial updating of that paper we might mention the more recent work on the problems associated with the Cauchy relations,[23] the asymmetries in the phase diagrams of CuNi,[24] α-CoFe,[25] and β-CuZn,[26] the ground state of CuPt,[27] and the influence of size effects[28] on ordering, or clustering, in binary alloys.

Quantitative models of the influence of static three-body interactions in binary alloys, based on the Ising model, have been introduced by Taggart and Tahir-Kheli[29] and Shirley and Wilkins.[30] In both of these works the binary systems was assumed to be described by a Hamiltonian of the form

$$\mathcal{H} = -\mu \sum_{g} S_g^z - \frac{1}{2}\sum_{g,p} I_2(gp) S_g^z S_p^z - \frac{1}{3}\sum_{g,p,f} I_3(gpf) S_g^z S_p^z S_f^z, \qquad (2.1)$$

where S_g^z is the z component of a spin-$\frac{1}{2}$ vector ($\hbar = 1$) at site g, and μ is an external field (chemical potential). $I_2(gp)$ is an irreducible two-body interaction between sites g and p, and $I_3(gpf)$ is an irreducible three-body interaction between sites g, p, and f. We could of course readily extend (2.1) to include other many-body interactions. For example a four-spin interaction in (2.1) with $I_3(gpf)$ and μ equal to zero would describe the ground state of CuPt,[27] and if we also set $I_2(gp)$ equal to zero we can model the order-disorder ferroelectric KH_2PO_4 (KDP).[31]

One proposal to explain the asymmetry of the phase diagrams of AuNi, AlZn, and other systems is to assume that, in the absence of explicit three-body interactions, the two-body interaction $I_2(gp)$ is concentration dependent.[32] However, this very idea is counter to the basic assumptions of the static model in which the interaction between two sites is assumed independent of the local environment, and hence independent of the concentration. However, the concept of a concentration-dependent two-body interaction was given some justification by the work of Taggart and Tahir-Kheli.[29] In the mean-field approximation they find a simple renormalization of the two-body interaction by the presence of the irreducible three-body interactions. This renormalization yields a concentration-dependent two-body interaction. As one goes beyond the mean-field approximation the influence of the three-body interactions on the two-body interactions becomes much more subtle and the concept of a concentration-dependent two-body interaction becomes weaker.

Using the formalism of Tahir-Kheli[19] and the results of Taggart and Tahir-Kheli[29] for binary alloys with three-body interactions, Vrijen *et al.*[24] have been able to explain recent diffuse neutron scattering results for clustering in CuNi systems. The use of the three-body interaction model was prompted by the failure of the two-body model in explaining the copper-rich side of the phase diagram (see also Ref. 23). In addition it is of interest to point out the work of Wood and Griffiths[33] who, using low temperature expansion methods, have shown that many-body interactions change the critical exponent β ($T \to T_c^-$), where

$$\langle S_g^z \rangle \sim (T_c - T)^{\beta}. \qquad (2.2)$$

Here $\langle\ \rangle$ represents the thermal average and for an alloy system $\langle S_g^z \rangle$ is proportional to the concentration.

Clapp[27] has shown that many-body interactions are necessary to determine the ground state of CuPt. This alloy possesses two completely equivalent ground state configurations, but only one is observed. In order to distinguish between ground states one has to assume the existence of at least four-body interactions. Interactions such as these, and three-body interactions, have been shown by Shirley[28] to be "caused" by size effects. In other words the strain energy caused by size effects produces an interaction that mimics many-body interactions.

Since one would believe that the many-body effects that have been observed in binary alloys would also be seen in ternary alloys, Taggart and Tahir-Kheli[34] extended the formalism of Ref. 29 to ternary systems. They calculated pair and triplet correlation functions to order $(T_c/T)^3$ and $(T_c/T)^2$, respectively. Within the mean-field approximation they found that the concept of an effective concentration-dependent pair-wise interaction is valid for the pair correlation function, but it is not for the triplet correlation function. In both cases in going beyond the mean-field approximation, and hence closer to the transition region, the influence of three-body interactions is far from trivial. As opposed to binary alloys, experimental indication of three-body interactions in ternary alloys appears to be lacking. Perhaps the most likely candidate for an analysis of three-body interactions in a ternary alloy would be to consider a binary alloy with vacancies as a ternary system. Regardless, any application of the results of Ref. 34 would require that we have an explicit expression for T_c, the transition temperature for ordering or clustering. The transition temperature in the mean-field, or Bragg-Williams, approximation has been calculated as a function of concentration and two-body interactions by a number of people.[7,10-13] However, calculations[34] that explicitly take into account the simplest many-body interaction, the three-body potential, have not derived the transition temperature. It is our purpose here to calculate the transition temperature for the ternary system with explicit static irreducible three-body interactions in the mean-field approximation using the correlation function approach.[17] First we will formulate the problem in Sec. III.

III. FORMULATION AND CORRELATION FUNCTIONS

We will assume that the three-component system can be adequately described by static interactions between atoms on a rigid lattice. We should recall, though, that for a binary system with pairwise interactions, the effects of atomic size disparity (strain effects) can cause pseudo-many-body effects to appear.[28] The lattice of N sites has N_ν atoms of type ν such that $\sum_\nu N_\nu = N$. The configurational energy can be written

$$H = \frac{1}{2!} \sum_{\substack{\nu,\rho \\ g,p}} V^{\nu\rho}(gp)\sigma_g^\nu \sigma_p^\rho + \frac{1}{3!} \sum_{\substack{\nu,\rho,\lambda \\ g,p,f}} W^{\nu\rho\lambda}(gpf)\sigma_g^\nu \sigma_p^\rho \sigma_f^\lambda , \tag{3.1}$$

where the sums over the labels ν, ρ, λ are over A, B, C, and the lattice sums are over all sites g, p, f.

We have introduced the usual occupation operator σ_g^ν in order to make the summations unrestricted, i.e.,

$$\sigma_g^\nu = \begin{cases} +1, & \text{if a } \nu \text{ atom is on site } g , \\ 0, & \text{otherwise.} \end{cases} \tag{3.2}$$

The static two-body interaction between sites g and p is given by $V^{\nu\rho}(gp)$ such that,

$$V^{\nu\rho}(gp) = V^{\rho\nu}(gp) = V^{\rho\nu}(pg) = V^{\nu\rho}(pg) , \tag{3.3a}$$

$$V^{\nu\rho}(gg) = V^{\rho\rho}(gg) = 0 . \tag{3.3b}$$

Likewise the static three-body interaction is assumed to have cyclic symmetry in both atomic and site labels, i.e.,

$$W^{\nu\nu\nu}(gpf) = W^{\nu\nu\nu}(fgp) = W^{\nu\nu\nu}(pfg) , \tag{3.4a}$$

$$W^{\nu\nu\rho}(gpf) = W^{\nu\nu\rho}(pgf) = W^{\nu\rho\nu}(gfp) , \tag{3.4b}$$

$$W^{\nu\rho\lambda}(gpf) = W^{\nu\lambda\rho}(gfp) , \tag{3.4c}$$

and

$$W^{\nu\rho\lambda}(gpf) = 0 , \tag{3.4d}$$

whenever any of the site indices g, p, f coincide.

Using the configurational Hamiltonian (3.1) exact correlation function relations can be derived.[15,34] In Ref. 34 this was done by transforming to a generalized Ising model by means of the spin transformations,

$$\sigma_g^A = \tfrac{1}{2}[(S_g^z)^2 + S_g^z] , \tag{3.5a}$$

$$\sigma_g^B = 1 - (S_g^z)^2 , \tag{3.5b}$$

$$\sigma_g^C = \tfrac{1}{2}[(S_g^z)^2 - S_g^z] , \tag{3.5c}$$

where S_g^z is the z component of a spin-1 vector ($\hbar = 1$) on site g. We thus obtain the generalized Ising Hamiltonian [compare (2.1)]

$$\mathcal{H}=-\mu_1\sum_{g} S_g^z-\mu_2\sum_{g}(S_g^z)^2-\sum_{g,p} I_1(gp)S_g^zS_p^z-\sum_{g,p} I_2(gp)S_g^z(S_p^z)^2-\sum_{g,p} I_3(gp)(S_g^z)^2(S_p^z)^2$$
$$-\sum_{g,p,f} I_4(gpf)S_g^zS_p^zS_f^z-\sum_{g,p,f} I_5(gpf)S_g^zS_p^z(S_f^z)^2-\sum_{g,p,f} I_6(gpf)(S_g^z)^2(S_p^z)^2S_f^z-\sum_{g,p,f} I_7(gpf)(S_g^z)^2(S_p^z)^2(S_f^z)^2\,, \qquad (3.6)$$

where the explicit relationships between the interactions $I_j(gp)$ and $I_j(gpf)$ with $V^{\nu\rho}(gp)$ and $W^{\nu\rho\lambda}(gpf)$ are given, for the sake of completeness, in the Appendix. The chemical potentials μ_j are determined such that the concentration is given by

$$m_\nu = N_\nu/N = \langle\sigma_g^\nu\rangle . \qquad (3.7)$$

The correlation function relations derived using the Hamiltonian (3.6) can then be expanded for $T > T_c$ in a self-consistent manner so that many-site correlation functions can be calculated to arbitrary order (in principle) in $\beta=(k_BT)^{-1}$ (k_B is the Boltzmann constant). The results of this calculation for the two-site correlation functions are[34]

$$\langle\sigma_g^A\sigma_p^C\rangle=(1-\delta_{gp})m_Am_C-\beta m_C(1-m_C)\Lambda_1(gp)$$
$$-\beta m_Am_C\Lambda_3(gp)+O(\beta^2) \qquad (3.8)$$

and

$$\langle\sigma_g^A\sigma_p^A\rangle=(m_A)^2+m_A(1-m_A)\delta_{gp}+\beta m_Am_C\Lambda_1(gp)$$
$$+\beta m_A(1-m_A)\Lambda_3(gp)+O(\beta^2) . \qquad (3.9)$$

Here we have used the notation ($j=1,3$)

$$\Lambda_j(gp)=m_A(1-m_A)U_j^A(gp)-m_Am_CU_j^C(gp)$$
$$+2m_A^2(1-m_A)\sum_f U_j^{AA}(gpf)$$
$$-2m_Am_C^2\sum_f U_j^{CC}(gpf)$$
$$+m_Am_C(1-2m_A)\sum_f U_j^{AC}(gpf)\,, \qquad (3.10)$$

and the parameters $U_j^\nu(gp)$ and $U_j^{\nu\rho}(gpf)$ are linear combinations of the interactions $I_j(gp)$ and $I_j(gpf)$ (see Appendix). We can obtain the other two-site correlation functions from (3.8) and (3.9) by interchanging the labels A, B, C, since we are using grand canonical averaging. We should note that we have to be careful during this interchange since the interactions $\Lambda_j(gp)$ are concentration dependent.

Having calculated the two-site correlation functions (it is straightforward to calculate many-site correlation functions using this technique) to the first order in β (the mean-field approximation) we can determine the Warren-Cowley[35] short-range-order (SRO) parameters $\alpha^{\nu\rho}(gp)$ from

$$m_\nu m_\rho\alpha^{\nu\rho}(gp)=m_\nu m_\rho-\langle\sigma_g^\nu\sigma_p^\rho\rangle\,, \qquad (3.11a)$$

$$m_\nu(1-m_\nu)\alpha^{\nu\nu}(gp)=\langle\sigma_g^\nu\sigma_p^\nu\rangle-m_\nu . \qquad (3.11b)$$

These parameters can be Fourier transformed

$$\alpha^{\nu\rho}(gp)=\frac{1}{N}\sum_{\vec{k}}\alpha^{\nu\rho}(\vec{k})e^{i\vec{k}\cdot(\vec{g}-\vec{p})}\,, \qquad (3.12a)$$

$$\alpha^{\nu\rho}(\vec{k})=\sum_{g,p}\alpha^{\nu\rho}(gp)e^{-i\vec{k}\cdot(\vec{g}-\vec{p})}\,, \qquad (3.12b)$$

and $\alpha^{\nu\rho}(\vec{k})$ can be shown to be proportional to the diffuse scattering intensity above T_c.[35] The problem now is to extract from $\alpha^{\nu\rho}(\vec{k})$ information about the transition temperature T_c. In Sec. IV we do this by using the correlation function approach to multicomponent alloys.

IV. CORRELATION FUNCTION APPROACH

Starting in the disordered regime ($T \gg T_c$) and lowering the temperature, the Fourier-transformed SRO parameter, or diffuse scattering intensity, $\alpha^{\nu\rho}(\vec{k})$, increases in magnitude until at some temperature T_c and wave vector $\vec{k}_c$ it becomes macroscopically large. This state of maximum amplitude in $\alpha^{\nu\rho}(\vec{k})$ defines the critical region at which the system goes from a disordered structure to an ordered one. (We admit both clustering and ordering in the "ordered" state in this approach.) Inspection of the two-site correlation function, (3.8) and (3.9), reveals no such singular behavior. In order to introduce this type of behavior into the problem Tahir-Kheli[36] has proposed that a self-consistent series inversion be performed. The results for the transition temperature, SRO parameters[36] and the interatomic potentials[19] generated by this procedure are quite promising for the binary alloy, and for this system the series-inversion technique presents no difficulties. However, for ternary and other multicomponent alloys the simple series inversion technique does not appear to give the proper results.[16,17] The solution to this difficulty is to recognize that for multicomponent systems we must use a matrix representation for the scattering intensities. The use of this matrix representation method has been called the correlation function approach to multicomponent alloys.[17] Its roots, as with the series inversion in binary alloys, are in the classical theory of critical fluctuations.[37] We refer the interested reader to Ref. 17, and here outline the salient features.

Using the Fourier-transformed two-site correlation functions given by (3.8) and (3.9) we can construct a correlation matrix $\underline{C}(\vec{k})$ whose elements are the SRO parameters $\alpha^{\nu\rho}(\vec{k})$. This matrix has the form

$$\underline{C}(\vec{k})=\begin{bmatrix} m_A(1-m_A)\alpha^{AA}(\vec{k}) & -m_Am_C\alpha^{AC}(\vec{k}) \\ -m_Am_C\alpha^{AC}(\vec{k}) & m_C(1-m_C)\alpha^{CC}(\vec{k}) \end{bmatrix}, \tag{4.1}$$

where only three independent SRO parameters are necessary to describe the ternary alloy (recall that $\sum_\nu \sigma_g^\nu = \sum_\nu m_\nu = 1$), and hence we need a matrix of dimension two. The numer of independent SRO parameters becomes more evident if we write $\underline{C}(\vec{k})$ in terms of the spin variables, i.e.,

$$\underline{C}(\vec{k})=\begin{bmatrix} \langle S_g^zS_p^z\rangle-\langle S_g^z\rangle^2 & \langle S_g^z(S_p^z)^2\rangle-\langle S_g^z\rangle\langle (S_p^z)^2\rangle \\ \langle S_g^z(S_p^z)^2\rangle-\langle S_g^z\rangle\langle (S_p^z)^2\rangle & \langle (S_g^z)^2(S_p^z)^2\rangle-\langle (S_g^z)^2\rangle^2 \end{bmatrix}_{(\vec{k})}, \tag{4.2}$$

where the subscript $(\vec{k})$ indicates that we have Fourier-transformed the components of $\underline{C}(\vec{k})$. Here we see that $\underline{C}(\vec{k})$ is composed of all two-site spin correlation functions of the vector space spanned by S_g^z and $(S_g^z)^2$.

Within the linear approximation in β (the mean-field approximation) we can explicitly write the matrix elements of (4.1), using the correlation functions given in (3.8) and (3.9). $\underline{C}(\vec{k})$ thus becomes

$$\underline{C}(\vec{k})=\underline{C}_0+\beta\underline{C}_1(\vec{k})+O(\beta^2), \tag{4.3}$$

where

$$\underline{C}_0=\begin{bmatrix} m_A(1-m_A) & -m_Am_C \\ -m_Am_C & m_C(1-m_C) \end{bmatrix} \tag{4.4}$$

and

$$\underline{C}_1(\vec{k})=\begin{bmatrix} m_Am_C\Lambda_1(\vec{k})+m_A(1-m_A)\Lambda_3(\vec{k}) & -m_C(1-m_C)\Lambda_1(\vec{k})-m_Am_C\Lambda_3(\vec{k}) \\ -m_A(1-m_A)\tilde{\Lambda}_1(\vec{k})-m_Am_C\tilde{\Lambda}_3(\vec{k}) & m_Am_C\tilde{\Lambda}_1(\vec{k})+m_C(1-m_C)\tilde{\Lambda}_3(\vec{k}) \end{bmatrix}. \tag{4.5}$$

The tilde on $\Lambda_j(\vec{k})$, i.e., $\tilde{\Lambda}_j(\vec{k})$, indicates that, in these concentration-dependent interactions, we interchange A and C (see Appendix). We can rewrite (4.3)

$$\underline{C}(\vec{k})=\underline{C}_0[\underline{1}+\beta\underline{\Gamma}_1(\vec{k})], \tag{4.6}$$

where

$$\underline{\Gamma}_1(\vec{k})=\underline{C}_0^{-1}\underline{C}_1(\vec{k}). \tag{4.7}$$

Within our linear, or mean-field, approximation and in the spirit of the series inversion procedure for the binary alloy, we can recast (4.6) in the form

$$\underline{C}(\vec{k})\simeq\underline{\Delta}[\underline{1}-\beta\underline{\Gamma}_1(\vec{k})]^{-1}. \tag{4.8}$$

The matrix $\underline{\Delta}$ is determined such that the total scattering intensity is conserved, i.e.,

$$\underline{C}_0=\frac{1}{N}\sum_{\vec{k}}\underline{C}(\vec{k}). \tag{4.9}$$

Looking at (4.8) we see that as $T\rightarrow T_c^+$ and $\vec{k}\rightarrow\vec{k}_c$ the magnitude of the scattering matrix will diverge, and hence,

$$|\underline{C}(\vec{k})|\rightarrow\infty \text{ when } T\rightarrow T_c,\ \vec{k}\rightarrow\vec{k}_c, \tag{4.10}$$

or

$$\frac{|\underline{\Delta}|}{|\underline{1}-\beta\underline{\Gamma}_1(\vec{k})|}\rightarrow\infty \text{ when } T\rightarrow T_c,\ \vec{k}\rightarrow\vec{k}_c. \tag{4.11}$$

The finiteness of $|\underline{\Delta}|$ implies that the critical condition is defined by the relation

$$|\underline{1}-\beta\underline{\Gamma}_1(\vec{k})|\rightarrow 0 \text{ when } T\rightarrow T_c,\ \vec{k}\rightarrow\vec{k}_c. \tag{4.12}$$

Thus (4.12) gives us an equation for the critical temperature β_c^{-1} in terms of the concentrations, and two- and three-body interactions of arbitrary strength and range. Tahir-Kheli[17] has used this procedure to calculate the transition temperature for ternary alloys with two-body potentials and has obtained agreement with the results derived previously, i.e., Refs. 10–13. Taggart[17] has used this technique to derive the transition temperature for a quaternary alloy with two-body interactions. We are now prepared to calculate β_c^{-1} from (4.12) for the ternary alloy with both two- and three-body interactions.

V. TRANSITION TEMPERATURE

The critical equation (4.12) is a quadratic equation in β_c^{-1} in the mean-field approximation for a three-component system, i.e.,

$$\beta_c^{-2}-\beta_c^{-1}\{[\Gamma_1(\vec{k}_c)]^{11}+[\Gamma_1(\vec{k}_c)]^{22}\} -\{[\Gamma_1(\vec{k}_c)]^{12}[\Gamma_1(\vec{k}_c)]^{21} -[\Gamma_1(\vec{k}_c)]^{11}[\Gamma_i(\vec{k}_c)]^{22}\}=0, \tag{5.1}$$

where $[\Gamma_1(\vec{k}_c)]^{ij}$ are the matrix elements of $\underline{\Gamma}_1(\vec{k}_c)$. The coefficients in (5.1) can be expressed explicitly in terms of two- and three-body interactions. (We will suppress the subscript on $\vec{k}$ for ease of notation.) We first define the two-body ordering interactions,

$$U^{\mu\nu}(gp) = V^{\mu\nu}(gp) - \tfrac{1}{2}[V^{\mu\mu}(gp) + V^{\nu\nu}(gp)] \,, \quad , \tag{5.2}$$

and the symmetrized three-body interactions,

$$\Omega^{\mu\mu\nu}(gpf) \equiv \tfrac{1}{3}[W^{\mu\mu\nu}(gpf) + W^{\mu\mu\nu}(gfp) + W^{\mu\mu\nu}(pfg)] \,, \tag{5.3a}$$

and

$$\begin{aligned}\Omega^{ABC}(gpf) \equiv \tfrac{1}{6}[&W^{ABC}(gpf) + W^{ABC}(gfp) \\ &+ W^{ABC}(pfg) + W^{ABC}(pgf) \\ &+ W^{ABC}(fgp) + W^{ABC}(fpg)] \,.\end{aligned} \tag{5.3b}$$

The solution to (5.1) can then be written

$$\begin{aligned}\beta_c^{-1} = \tfrac{1}{2}[J^{AB}(\vec{k}) + J^{AC}(\vec{k}) + J^{BC}(\vec{k})] \pm \tfrac{1}{2}\Bigg\{&[J^{AB}(\vec{k}) + J^{AC}(\vec{k}) + J^{BC}(\vec{k})]^2 \\ &+ m_A m_B m_C \Bigg[\left(\frac{J^{AB}(\vec{k})}{m_A m_B} + \frac{J^{AC}(\vec{k})}{m_A m_C} + \frac{J^{BC}(\vec{k})}{m_B m_C}\right)^2 \\ &- 4\left(\frac{J^{AB}(\vec{k})J^{AC}(\vec{k})}{m_A^2 m_B m_C} + \frac{J^{AB}(\vec{k})J^{BC}(\vec{k})}{m_A m_B^2 m_C} + \frac{J^{AC}(\vec{k})J^{BC}(\vec{k})}{m_A m_B m_C^2}\right)\Bigg]\Bigg\}^{1/2} \,,\end{aligned} \tag{5.4}$$

where we have defined the effective concentration dependent two-body interactions as

$$\begin{aligned}J^{\mu\nu}(\vec{k}) = &-2m_\mu m_\nu U^{\mu\nu}(\vec{k}) + m_\mu^2 m_\nu W^{\mu\mu\mu}(\vec{k}) \\ &+ m_\mu m_\nu^2 W^{\nu\nu\nu}(\vec{k}) + m_\mu m_\nu(1 - 3m_\mu)\Omega^{\mu\mu\nu}(\vec{k}) \\ &+ m_\mu m_\nu(1 - 3m_\nu)\Omega^{\nu\nu\mu}(\vec{k}) - 2m_\mu m_\nu m_\rho \Omega^{\mu\nu\rho}(\vec{k}) \,.\end{aligned} \tag{5.5}$$

The behavior of β_c^{-1} as a function of concentration and interaction strength is not readily apparent from (5.4). However, we can gain some insight into the behavior of β_c^{-1} by investigating some appropriate limits. It should be stressed, though, that our result is valid for arbitrary concentration, arbitrary range of interactions, and arbitrary strengths of interactions.

The first interesting limit would be the binary alloy limit ($m_C \rightarrow 0$), which should supplement the results of Taggart and Tahir-Kheli[29] who used an expansion to β^3 and Shirley and Wilkins[30] who used a z^{-1} expansion (z equals the number of nearest neighbors). In this limit (5.4) yields,

$$\begin{aligned}\beta_c^{-1} = -2m_A m_B U^{AB}(\vec{k}) + [&m_A^2(1 - m_A)W^{AAA}(\vec{k}) + m_B^2(1 - m_B)W^{BBB}(\vec{k}) \\ &+ m_A m_B(1 - 3m_A)\Omega^{AAB}(\vec{k}) + m_A m_B(1 - 3m_B)\Omega^{BBA}(\vec{k})] \,.\end{aligned} \tag{5.6}$$

Setting the three-body potentials to zero yields immediately the simple binary alloy result with only two-body interactions, i.e.,

$$\beta_c^{-1} = -2m_A m_B U^{AB}(\vec{k}) \,. \tag{5.7}$$

We also note that, in this mean-field approximation, we could represent the interaction energy in (5.6) by an effective concentration-dependent two-body interaction. As we pointed out in Ref. 29 this gives some justification for using concentration-dependent interactions in order to describe the phase diagram of AlZn (Ref. 32) for instance. However, as was also pointed out in Ref. 29 this concept of a simple concentration-dependent effective two-body interaction loses validity as one considers higher-order terms in the β expansion, and hence more accurate approximations to the transition region. In that case one would have to take explicit account of the lowest order many-particle interaction, the three-body interaction as we have done here.

If we look at (5.6) for small deviations from stoichiometry, i.e., $m_A = \frac{1}{2} + x, m_B = \frac{1}{2} - x$ ($x \ll 1$), and retain only terms linear in x, we obtain

$$\beta_c(x)^{-1} = -\tfrac{1}{2}U^{AB}(\vec{k}) + \tfrac{1}{8}\times[W^{AAA}(\vec{k}) + W^{BBB}(\vec{k}) - \Omega^{AAB}(\vec{k}) - \Omega^{BBA}(\vec{k})] + \tfrac{1}{4}x\,[W^{AAA}(\vec{k}) - W^{BBB}(\vec{k}) - 3\Omega^{AAB}(\vec{k}) + 3\Omega^{BBA}(\vec{k})] + O(x^2). \tag{5.8}$$

The asymmetry of the phase diagram is apparent from (5.8) as one moves away from the equicomposition binary alloy. This becomes even more apparent if we look at the symmetric alloy,[38] i.e., $x \rightarrow -x$, and calculate the difference in transition temperatures,

$$\beta_c(x)^{-1} - \beta_c(-x)^{-1} = \tfrac{1}{2}x\,[W^{AAA}(\vec{k}) - W^{BBB}(\vec{k}) - 3\Omega^{AAB}(\vec{k}) + 3\Omega^{BBA}(\vec{k})]\,. \tag{5.9}$$

Thus deviations from symmetry about the equicomposition alloy would give, in this static model, an indication of the effects of many-body interactions. Likewise complete symmetry would indicate the absence of these interactions.[39] Since we do not expect the properties of the ternary alloy to be appreciably different than those of the binary alloy, we would expect similar types of behavior in the ternary system.

For the ternary alloy we first look at the two-body interaction limit. Thus setting the three-body interactions to zero, we obtain

$$\begin{aligned}\beta_c^{-1} = {} & -m_A m_B U^{AB}(\vec{k}) - m_A m_C U^{AC}(\vec{k}) - m_B m_C U^{BC}(\vec{k}) \\ & \pm(\,[m_A m_B U^{AB}(\vec{k}) + m_A m_C U^{AC}(\vec{k}) + m_B m_C U^{BC}(\vec{k})]^2 + m_A m_B m_C\{[U^{AB}(\vec{k}) + U^{AC}(\vec{k}) + U^{BC}(\vec{k})]^2 \\ & - 4[U^{AB}(\vec{k})U^{AC}(\vec{k}) + U^{AB}(\vec{k})U^{BC}(\vec{k}) + U^{AC}(\vec{k})U^{BC}(\vec{k})]\}\,)^{1/2}\,,\end{aligned} \tag{5.10}$$

which is the same expression calculated by numerous other authors.[7,10-13] In the limit of small concentrations of m_C and equicompositions of A and B, i.e., $m_A = m_B = \frac{1}{2}(1 - m_C), m_C \ll 1$, (5.10) becomes

$$\beta_c^{-1} = -\tfrac{1}{2}U^{AB}(\vec{k}) \times \left\{1 - m_C\left[1 - \left(\frac{U^{AC}(\vec{k}) - U^{BC}(\vec{k})}{U^{AB}(\vec{k})}\right)^2\right]\right\} + O(m_C^2)\,, \tag{5.11}$$

which has been calculated previously by Prigogine[9] and Meijering,[7] as well as Tahir-Kheli.[13,40]

In order to examine the influence of three-body interactions we will also look at (5.4) in the limit that $m_C \ll 1$ and $m_A = m_B = (1 - m_C)/2$. We can then compare this result with (5.11). Keeping only terms linear in m_C we first define the binary transition temperature by means of (5.6), i.e.,

$$\beta_c(AB)^{-1} \equiv -\tfrac{1}{2}U^{AB}(\vec{k}) + \tfrac{1}{8}[W^{AAA}(\vec{k}) + W^{BBB}(\vec{k}) - \Omega^{AAB}(\vec{k}) - \Omega^{BBA}(\vec{k})] \tag{5.12}$$

and thus write for the ternary transition temperature,

$$\begin{aligned}\beta_c^{-1} = \beta_c(AB)^{-1}\,[1 - \tfrac{1}{4}m_C\beta_c(AB)^2(U^{AB}(\vec{k})^2 - [U^{AC}(\vec{k}) - U^{BC}(\vec{k})]^2 \\ + \beta_c(AB)^{-1}\tfrac{1}{16}[W^{AAA}(\vec{k}) + W^{BBB}(\vec{k}) - 5\Omega^{AAB}(\vec{k}) - 5\Omega^{BBA}(\vec{k}) + 2\Omega^{AAC}(\vec{k}) \\ + 2\Omega^{BBC}(\vec{k}) + 2\Omega^{ABC}(\vec{k}) - 4\Omega^{CCA}(\vec{k}) - 4\Omega^{CCB}(\vec{k})] \\ - U^{AC}(\vec{k})[W^{BBB}(\vec{k}) + \Omega^{AAC}(\vec{k}) - \Omega^{AAB}(\vec{k}) - \Omega^{BBC}(\vec{k})] \\ - U^{BC}(\vec{k})[W^{AAA}(\vec{k}) + \Omega^{BBC}(\vec{k}) - \Omega^{AAC}(\vec{k}) - \Omega^{BBA}(\vec{k})] \\ - U^{AB}(\vec{k})[\Omega^{BBA}(\vec{k}) - \Omega^{BBC}(\vec{k}) + \Omega^{AAB}(\vec{k}) - \Omega^{AAC}(\vec{k}) + \Omega^{CCA}(\vec{k}) + \Omega^{CCB}(\vec{k}) - 2\Omega^{ABC}(\vec{k})] \\ + \tfrac{1}{4}\{W^{AAA}(\vec{k})W^{BBB}(\vec{k}) + W^{BBB}(\vec{k})\Omega^{AAB}(\vec{k}) + W^{AAA}(\vec{k})\Omega^{CCA}(\vec{k}) \\ + W^{AAA}(\vec{k})\Omega^{BBA}(\vec{k}) + W^{BBB}(\vec{k})\Omega^{CCB}(\vec{k}) + W^{AAA}(\vec{k})\Omega^{CCB}(\vec{k}) \\ - 2W^{AAA}(\vec{k})\Omega^{BBC}(\vec{k}) - 2W^{BBB}(\vec{k})\Omega^{AAC}(\vec{k}) \\ + W^{BBB}(\vec{k})\Omega^{CCA}(\vec{k}) - 2[W^{AAA}(\vec{k}) + W^{BBB}(\vec{k})]\Omega^{ABC}(\vec{k}) + 2\Omega^{AAB}(\vec{k})\Omega^{AAC}(\vec{k}) \\ + 2\Omega^{BBA}(\vec{k})\Omega^{BBC}(\vec{k}) - \Omega^{AAB}(\vec{k})\Omega^{BBA}(\vec{k}) - \Omega^{BBA}(\vec{k})\Omega^{CCB}(\vec{k}) \\ - \Omega^{AAB}(\vec{k})\Omega^{CCA}(\vec{k}) - \Omega^{AAB}(\vec{k})\Omega^{CCB}(\vec{k}) - \Omega^{BBA}(\vec{k})\Omega^{CCA}(\vec{k}) \\ + 2\Omega^{AAC}(\vec{k})\Omega^{BBC}(\vec{k}) + 2[\Omega^{AAB}(\vec{k}) + \Omega^{BBA}(\vec{k})]\Omega^{ABC}(\vec{k}) \\ - \Omega^{AAB}(\vec{k})^2 - \Omega^{BBA}(\vec{k})^2 - \Omega^{AAC}(\vec{k})^2 - \Omega^{BBC}(\vec{k})^2\})] + O(m_C^2)\,.\end{aligned} \tag{5.13}$$

From (5.13) we see that if the three-body interactions vanish we retrieve (5.11). As would be expected in this particular limit the transition-temperature behavior is dominated by the binary transition. However, we also see that even in this simple limit the influence of three-body potentials is far from trivial if we desire to explicitly take them into account.

VI. SUMMARY

Using the correlation function approach to multicomponent alloys we have calculated the transition temperature in the mean-field approximation for a ternary alloy having both two- and three-body potentials. Our result is valid for arbitrary concentrations, arbitrary range, and strength of interactions, and should apply to both clustering and ordering systems. We see that within the mean-field approximation we can represent the interactions by effective concentration-dependent two-body interactions. That is to say, the interactions $J^{\mu\nu}(\vec{k})$ are concentration dependent.

Using high temperature expansion techniques, if we went to higher order approximations in β we would systematically include multisite correlation functions to higher orders in our calculation of the pair correlation function. Thus our results should describe a smaller neighborhood of the critical point. However, as this point is approached the three-body interactions will manifest themselves in such a manner that a simple linear renormalization, i.e., $J^{\mu\nu}(\vec{k})$, of the two-body potential is no longer possible. This linear renormalization is characteristic of the mean-field approximation.

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APPENDIX

The relations between the interactions in the spin representation and the occupation-operator representation are given by

$$I_1(gp) = -\tfrac{1}{8}\left[V^{AA}(gp) + V^{BB}(gp)\right] + \frac{1}{8}\sum_f \left[W^{ABC}(gfp) + W^{ABC}(pfg) - W^{AAB}(gpf) - W^{CCB}(gpf)\right], \tag{A.1a}$$

$$\begin{aligned} I_2(gp) = {} & \tfrac{1}{2}\left[V^{AB}(gp) - V^{BC}(gp)\right] + \tfrac{1}{4}\left[V^{CC}(gp) - V^{AA}(gp)\right] + \frac{1}{2}\sum_f \left[W^{BBA}(gfp) - W^{BBC}(gfp)\right] \\ & + \frac{1}{4}\sum_f \left[W^{ABC}(gfp) - W^{ABC}(pfg) + W^{CCB}(gpf) - W^{AAB}(gpf)\right], \end{aligned} \tag{A.1b}$$

$$\begin{aligned} I_3(gp) = {} & \tfrac{1}{2}\left[V^{AB}(gp) + V^{BC}(gp) - V^{BB}(gp)\right] - \tfrac{1}{8}\left[V^{AA}(gp) + V^{CC}(gp) + 2V^{AC}(gp)\right] \\ & + \frac{1}{4}\sum_f \left[W^{BBA}(gfp) + W^{BBA}(pfg) + W^{BBC}(gfp) + W^{BBC}(pfg) - 2W^{BBB}(gpf)\right] \\ & - \frac{1}{8}\sum_f \left[W^{ABC}(gfp) + W^{ABC}(pfg) + W^{AAB}(gpf) + W^{CCB}(gpf)\right], \end{aligned} \tag{A.1c}$$

$$\begin{aligned} I_4(gpf) = {} & \tfrac{1}{48}\left[W^{AAC}(gpf) - W^{CCA}(gpf) + W^{AAC}(gfp) + W^{AAC}(pfg) - W^{CCA}(gfp)\right. \\ & \left. - W^{CCA}(pfg) + W^{CCC}(gpf) - W^{AAA}(gpf)\right], \end{aligned} \tag{A.1d}$$

$$\begin{aligned} I_5(gpf) = {} & \tfrac{1}{16}\left[2W^{AAB}(gpf) + 2W^{CCB}(gpf) - 2W^{ABC}(gfp) - 2W^{ABC}(pfg) - W^{AAA}(gpf)\right. \\ & - W^{CCC}(gpf) - W^{AAC}(gpf) + W^{AAC}(gfp) \\ & \left. + W^{AAC}(pfg) - W^{CCA}(gpf) + W^{CCA}(gfp) + W^{CCA}(pfg)\right], \end{aligned} \tag{A.1e}$$

$$\begin{aligned} I_6(gpf) = {} & \tfrac{1}{16}\left[W^{CCC}(gpf) - W^{AAA}(gpf) + W^{AAC}(gpf) - W^{AAC}(gfp) - W^{AAC}(pfg) - W^{CCA}(gpf)\right. \\ & + W^{CCA}(gfp) + W^{CCA}(pfg) - 4W^{BBA}(gpf) - 4W^{BBC}(gpf) + 2W^{AAB}(gfp) \\ & + 2W^{AAB}(pfg) - 2W^{CCB}(gfp) - 2W^{CCB}(pfg) \\ & \left. + 2W^{ABC}(fgp) + 2W^{ABC}(fpg) - 2W^{ABC}(gpf) - 2W^{ABC}(pgf)\right], \end{aligned} \tag{A.1f}$$

and,

$$\begin{aligned} I_7(gpf) = \tfrac{1}{48}[&8W^{BBB}(gpf) - W^{AAA}(gpf) - W^{CCC}(gpf) - W^{AAC}(gpf) - W^{AAC}(gfp) \\ &- W^{AAC}(pfg) - W^{CCA}(gpf) \\ &- W^{CCA}(gfp) - W^{CCA}(pfg) - 4W^{BBA}(gpf) - 4W^{BBA}(gfp) - 4W^{BBA}(pfg) \\ &- 4W^{BBC}(gpf) - 4W^{BBC}(gfp) \\ &- 4W^{BBC}(pfg) + 2W^{ABC}(gpf) + 2W^{ABC}(gfp) + 2W^{ABC}(pgf) + 2W^{ABC}(pfg) \\ &+ 2W^{ABC}(fgp) + 2W^{ABC}(fpg) + 2W^{AAB}(gpf) + 2W^{AAB}(gfp) + 2W^{AAB}(pfg) \\ &+ 2W^{CCB}(gfp) + 2W^{CCB}(gpf) + 2W^{CCB}(pfg)]\,. \end{aligned} \tag{A.1g}$$

The parameters $U_j^{\nu}(gp)$ and $U_j^{\nu\rho}(gpf), j=1,3$, are given in terms of $I_j(gp)$ and $I_j(gpf)$ by

$$U_1^{A}(gp) = 2I_1(gp) - 2I_3(gp)\,, \tag{A.2a}$$

$$U_1^{C}(gp) = 2I_2(gp) - 2I_1(gp) - 2I_3(gp)\,, \tag{A.2b}$$

$$U_1^{AA}(gpf) = 3I_4(gpf) + I_5(gpf) - I_6(gpf) - 3I_7(gpf)\,, \tag{A.2c}$$

$$U_1^{CC}(gpf) = 3I_4(gpf) - 3I_5(gpf) + 3I_6(gpf) - 3I_7(gpf)\,, \tag{A.2d}$$

$$U_1^{AC}(gpf) = -6I_4(gpf) + 2I_5(gpf) + 2I_6(gpf) - 6I_7(gpf)\,, \tag{A.2e}$$

$$U_3^{A}(gp) = -\tilde{U}_1^{C}(gp)\,, \tag{A.3a}$$

$$U_3^{C}(gp) = -U_1^{A}(gp)\,, \tag{A.3b}$$

$$U_3^{AA}(gpf) = -\tilde{U}_1^{CC}(gpf)\,, \tag{A.3c}$$

$$U_3^{CC}(gpf) = -\tilde{U}_1^{AA}(gpf)\,, \tag{A.3d}$$

$$U_3^{AC}(gpf) = -\tilde{U}_1^{AC}(gpf)\,, \tag{A.3e}$$

where the tilde implies that we interchange A and C.

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[*] Permanent address: Dept. of Physics, Virginia Commonwealth Univ., Richmond, Va. 23284

[1] H. Sato, *Physical Chemistry: An Advanced Treatise*, Vol. 10, edited by W. Jost (Academic, New York, 1970).

[2] Ø. Fischer, Appl. Phys. 16, 1 (1978).

[3] S. Iijima and J. M. Cowley, J. Phys. (Paris) Colloq. 38, C7-135 (1977).

[4] G. Dublon, U. Atzmony, and M. P. Dariel, J. Magn. Magn. Mater. 7, 131 (1978).

[5] H. Kasano, J. Appl. Phys. 49, 4021 (1978).

[6] Y. Murakami, N. Nakanishi, and S. Kachi, Acta Metall. 19, 93 (1971).

[7] J. L. Meijering, Philips Res. Rep. 5, 333 (1950); 6, 183 (1951).

[8] K. F. Wojciechowski, Acta Metall. 6, 396 (1958).

[9] I. Prigogine, Bull. Soc. Chim. Belg. 52, 115 (1943); S. Matsuda, J. Phys. Soc. Jpn. 8, 20 (1953).

[10] Y. Murakami, S. Kachi, N. Nakanishi, and H. Takehara, Acta Metall. 19, 97 (1971).

[11] D. de Fontaine, J. Phys. Chem. Solids 33, 297 (1972); 34, 1285 (1973).

[12] V. I. Ryzhkov, Fiz. Met. Metalloved. 41, 7 (1976).

[13] R. A. Tahir-Kheli, Phys. Status Solidi B 79, 93 (1977).

[14] A. G. Khachaturyan, Prog. Mater. Sci, 22, 1 (1978).

[15] R. A. Tahir-Kheli, Phys. Rev. 169, 517 (1968); G. B. Taggart and R. A. Tahir-Kheli, Phys. Rev. B3 1059 (1971).

[16] G. B. Taggart, J. Phys. Chem. Solids 33, 2287 (1972); D. de Fontaine, J. Appl. Cryst. 4, 15 (1971).

[17] R. A. Tahir-Kheli (unpublished); G. B. Taggart, Phys. Rev. B (to be published).

[18] D. M. Burley, *Phase Transitions and Critical Phenomena,* Vol. 2, edited by C. Domb and M. S. Green (Academic, New York, 1972), Chap. 9.

[19] D.-H. Wu and R. A. Tahir-Kheli, J. Phys. Soc. Jpn. 31, 641 (1971).

[20] N. S. Golosov and A. M. Tolstik, J. Phys. Chem. Solids 35 1575 (1974); R. Kikuchi, Acta Metall. 25, 195 (1977); R. Kikuchi, D. de Fontaine, M. Murakami, and T. Nakamura, *ibid.* 25, 207 (1977).

[21] G. B. Taggart (unpublished).

[22] P. C. Clapp, *Ordered Alloys*, edited by B. H. Kear, C. T. Sims, N. S. Stoloff, and J. H. Westbrook (Claitors, Baton Rouge, 1970), p. 25.

[23] C. S. G. Cousins, J. Phys. F 3, 1915 (1973); J. W. Martin, J. Phys. C 8, 2837 (1975); 8, 2858 (1975).

[24] J. Vrijen, Ph.D. dissertation (Rijksuniversiteit, Utrecht, 1977) (unpublished); J. Vrijen, E. W. van Royen, D. W. Hoffman, and S. Radelaar, J. Phys. Paris Colloq. 38, C7-187 (1977); J. Vrijen and S. Radelaar, Phys. Rev. B17, 409 (1978).

[25] F. A. Oyedale and M. F. Collins, Phys. Rev. B16, 3208 (1977).

[26] D. Yoon and A. Bienenstock, Phys. Rev. 170, 631 (1968); D. Yoon and R. Jeffery, J. Phys. Chem. Solids

31, 2635 (1970).

[27] P. C. Clapp, Acta Metall. 22, 563 (1974).

[28] C. G. Shirley, Phys. Rev. B 10, 1149 (1974).

[29] G. B. Taggart and R. A. Tahir-Kheli, Prog. Theor. Phys. 46, 1690 (1971).

[30] C. G. Shirley and S. W. Wilkins, Phys. Rev. B 6 1252 (1972); 16, 3484 (1977).

[31] M. E. Lines and A. M. Glass, *Principles and Applications of Ferroelectrics and Related Materials* (Oxford, New York, 1977), p. 293.

[32] M. Hillert, *Metallic Solid Solutions*, edited by J. Friedel and A. Guinier (Benjamin, New York, 1963).

[33] D. W. Wood and H. P. Griffiths, J. Phys. C 7, L54 (1974).

[34] G. B. Taggart and R. A. Tahir-Kheli, Prog. Theor. Phys. 47, 370 (1972).

[35] B. E. Warren, *X-Ray Diffraction* (Addison-Wesley, Reading, Mass., 1969); J. M. Cowley, *Diffraction Physics* (North-Holland, Amsterdam, 1975).

[36] R. A. Tahir-Kheli, Phys. Rev. 188, 1142 (1969); G. B. Taggart and R. A. Tahir-Kheli, Physica (Utr.) 68, 93 (1973).

[37] A. Munster, *Fluctuation Phenomena in Solids*, edited by R. E. Burgess (Academic, New York, 1965).

[38] W. H. Kleiner, J. Chem. Phys. 36, 76 (1962).

[39] R. A. Tahir-Kheli, Phys. Lett. A25, 641 (1967); P. C. Clapp, Phys. Rev. 164, 1018 (1967).

[40] Equation (5.13) of Ref. 13 has been transcribed improperly; however, the original agrees with our Eq. (5.12). [R. A. Tahir-Kheli (private communication).]