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Theory of Disordered Ternary Alloys at Intermediate Temperatures

G. Bruce Taggart[*,†]
Department of Physics, Temple University, Philadelphia, Pennsylvania 19122
and
Raza A. Tahir-Kheli
Laboratoire de Physique des Solides,[‡] *Faculté des Sciences, 91-Orsay, France and*
Department of Physics, Temple University,[§] *Philadelphia, Pennsylvania* 19122

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Expressions for the short-range-order parameters for disordered ternary alloys are given as functions of the system composition and the static two-body configurational potentials of arbitrary range. These expressions are exact to the order $(T_c/T)^3$, where T_c is of the order of the highest temperature at which the system is expected to have any long-range order. A procedure for the computation of this temperature T_c is outlined. It is noted that the present approximation removes an obvious inadequacy of an earlier linear-approximation treatment.

I. INTRODUCTION

Exact formal solutions can be derived[1] for the short-range-order (SRO) parameters for a disordered ternary alloy, with arbitrary compositions and nondynamical pair-wise interatomic potentials of arbitrary range. To do explicit calculations, a suitable high-temperature expansion scheme can be devised. In Ref. 1, hereafter denoted as I, the series expansion for the SRO parameters was truncated at the dominant temperature-dependent term. Such a linear approximation can be expected to yield adequate results in the limit of elevated temperatures, i.e., $T \gg T_c$, where T_c is the highest temperature at which the system is capable of showing any long-range spatial order.

The linear approximation discussed in I suffers from two major inadequacies. First, its accuracy at intermediate temperatures, i.e., $T_c < T < 2T_c$, is expected to be poor. Second, the estimates of T_c obtained within the linear-approximation scheme, have some unphysical features whenever in the ternary system the concentration of one of the components is close to 50%. Moreover, in the limit that the relative concentration of one of the components is vanishing, i.e., when the system becomes a binary alloy, it has recently been shown[2] that the predictions of the linear approximation regarding the dependence of T_c upon the concentration are in serious disagreement with experimental observations on the order-disorder temperature of the copper-gold binary system. In contrast to this manifest failure of the linear approximation, the predictions of the improved cubic approximation are found to be[2] in very reasonable agreement with experiment.

To remedy these deficiencies, in the present paper we present a more accurate calculation of the SRO parameters. In terms of the appropriate high-temperature series expansion, these results are exact to the third order in the ratio T_c/T. As such, they are an appropriate generalization of the corresponding binary results[2] to the case of ternary systems.

II. SRO PARAMETERS

In I we described a disordered ternary alloy with a total of N atoms of which N^A, N^B, and $N^C = N - N^A - N^B$ are of types A, B, and C, respectively. These atoms are distributed on a regular isotropic rigid lattice consisting of N sites. The configurational interaction is a sum of two-body interactions, i.e.,

$$H = H^{AA} + H^{BB} + H^{CC} + H^{AB} + H^{AC} + H^{BC} , \qquad (2.1a)$$

where

$$H^{\lambda\lambda} = \tfrac{1}{2} \sum_{i,j} V^{\lambda\lambda}(ij) \sigma_i^\lambda \sigma_j^\lambda , \qquad (2.1b)$$

$$H^{\lambda\nu}=\tfrac{1}{2}\sum_{i,j}V^{\lambda\nu}(ij)(\sigma_i^\lambda\sigma_j^\nu+\sigma_i^\nu\sigma_j^\lambda)\,,\quad \lambda\neq\nu \qquad (2.1\text{c})$$

$$V^{\lambda\nu}(ij)=V^{\nu\lambda}(ij)=V^{\nu\lambda}(ji)=V^{\lambda\nu}(ji)\;, \qquad (2.1\text{d})$$

$$V^{\lambda\nu}(ii)\equiv V^{\lambda\lambda}(ii)\equiv 0\,,\quad \lambda,\ \nu=A,\ B,\ C\;. \qquad (2.1\text{e})$$

Here the sums over i and j are unrestricted and extend over all the atoms in the lattice. This is made possible by the use of the occupation operators σ_i^λ which are defined as

$$\begin{aligned}\sigma_i^\lambda&=+1\quad \text{if the } i\text{th lattice position is occupied}\\ &\qquad\quad\ \text{by an atom of type } \lambda\,,\\ &=0\qquad \text{otherwise}\;. \end{aligned}\qquad (2.1\text{f})$$

Transforming to the subspace of spin 1 (in units such that $\hbar=1$) by the way of the relations

$$\sigma_i^A=\tfrac{1}{2}\left[(S_i^z)^2+S_i^z\right]\,, \qquad (2.2\text{a})$$

$$\sigma_i^B=1-(S_i^z)^2\,, \qquad (2.2\text{b})$$

$$\sigma_i^C=\tfrac{1}{2}\left[(S_i^z)^2-S_i^z\right] \qquad (2.2\text{c})$$

(refer to Appendix C for the details of this transformation), we obtain the following system Hamiltonian from (2.1):

$$H=-\frac{1}{2}\sum_{i,j}\left[I_1(ij)S_i^zS_j^z+I_2(ij)(S_i^z)^2(S_j^z)^2+I_3(ij)(S_i^z)^2S_j^z\right]$$
$$-\,\mu\sum_i S_i^z-\rho\sum_i (S_i^z)^2\,, \qquad (2.3\text{a})$$

$$I_1(ij)=\tfrac{1}{4}\left[2V^{AC}(ij)-V^{CC}(ij)-V^{AA}(ij)\right]\,, \qquad (2.3\text{b})$$

$$I_2(ij)=V^{AB}(ij)+V^{BC}(ij)-V^{BB}(ij)$$
$$-\tfrac{1}{4}\left[V^{AA}(ij)+V^{CC}(ij)+2V^{AC}(ij)\right]\,, \qquad (2.3\text{c})$$

$$I_3(ij)=V^{AB}(ij)-V^{BC}(ij)+\tfrac{1}{2}\left[V^{CC}(ij)-V^{AA}(ij)\right]\;. \qquad (2.3\text{d})$$

The chemical potentials μ and ρ are computed from

$$\langle N^\lambda\rangle=N^\lambda \qquad (2.4)$$

(angular brackets denote the usual thermal average).

As shown in I, the Hamiltonian (2.3) can be solved for exact formal solutions of the spin correlation functions $\langle S_g^z[\cdots]\rangle$, where $[\cdots]$ refers to any S^z operator *not* referring to lattice site g. These correlation functions are related to the SRO parameters $\alpha^{\lambda\nu}(\overrightarrow{\mathrm{gp}})$ through their dependence on the occupation-operator correlation functions, i.e.,

$$\alpha^{\lambda\nu}(\overrightarrow{\mathrm{gp}})=(m^\lambda m^\nu-\langle\sigma_g^\lambda\sigma_p^\nu\rangle)/m^\lambda m^\nu\,, \qquad (2.5)$$

where m^λ is the relative concentration of the λ atoms,

$$m^\lambda=N^\lambda/N=\langle\sigma_g^\lambda\rangle\;. \qquad (2.6)$$

It is, therefore, convenient to recast the formal results derived in I for the spin correlation functions in terms of the occupation-operator representation. This is readily done by first inverting Eq. (2.2a), i.e.,

$$S_i^z=\sigma_i^A-\sigma_i^C\,, \qquad (2.7\text{a})$$

$$(S_i^z)^2=\sigma_i^A+\sigma_i^C\,, \qquad (2.7\text{b})$$

and using these to reexpress Eqs. (3.6), (3.7), and (3.15)–(3.18) of I, in terms of the A and C occupation operators (note that $\sigma_i^B=1-\sigma_i^A-\sigma_i^C$). We get

$$\langle[\cdots]\sigma_g^A\rangle=\langle[\cdots]\varphi(3)(1-\sigma_g^C)\rangle\,, \qquad (2.8\text{a})$$

$$\langle[1-\varphi(1)-\varphi(3)][\cdots]\rangle$$
$$=\langle[1-\varphi(3)]\sigma_g^C[\cdots]\rangle-\langle\varphi(1)\sigma_g^A[\cdots]\rangle\,, \qquad (2.8\text{b})$$

where we have used the notation

$$\varphi(n)=(e^{-\beta E^{(n)}(g)}+1)^{-1}\,,\quad n=1,\ 3 \qquad (2.9\text{a})$$

$$E^{(1)}(g)=\mu-\rho-\textstyle\sum_f\left[W(gf)\sigma_f^A+2U^{BC}(gf)\,\sigma_f^C\right]\,, \qquad (2.9\text{b})$$

$$E^{(3)}(gf)=\mu+\rho+\textstyle\sum_f\left[2U^{AB}(gf)\,\sigma_f^A+W(gf)\,\sigma_f^C\right]\,, \qquad (2.9\text{c})$$

$$U^{\lambda\nu}(gf)=\tfrac{1}{2}\left\{2V^{\lambda\nu}(gf)-\left[V^{\lambda\lambda}(gf)+V^{\nu\nu}(gf)\right]\right\}\,, \qquad (2.9\text{d})$$

$$W(gf)=U^{AB}(gf)+U^{BC}(gf)-U^{AC}(gf)\;. \qquad (2.9\text{e})$$

As discussed in I, above T_c the functions $\varphi(1)$ and $\varphi(3)$ can be expanded in powers of T_c/T. The appropriate high-temperature expansions are

$$\varphi(1)=X(0)+\sum_{n=1}^{\infty}X(n)[\beta\tilde{E}^{(1)}(g)]^n\,, \qquad (2.10\text{a})$$

$$\varphi(3)=Y(0)+\sum_{n=1}^{\infty}Y(n)[\beta\tilde{E}^{(3)}(g)]^n\,, \qquad (2.10\text{b})$$

where

$$\tilde{E}^{(1)}(g)=E^{(1)}(g)-\mu+\rho\,, \qquad (2.11\text{a})$$

$$\tilde{E}^{(3)}(g)=E^{(3)}(g)-\mu-\rho\;. \qquad (2.11\text{b})$$

Here the coefficients $X(n)$ and $Y(n)$ are functions only of $X(0)$ and $Y(0)$,

$$X(0)=(1+e^{-\beta(\mu-\rho)})^{-1}\,, \qquad (2.12\text{a})$$

$$Y(0)=(1+e^{-\beta(\mu+\rho)})^{-1}\;. \qquad (2.12\text{b})$$

For example, we have

$$X(1)=X(0)\,[1-X(0)]\,, \qquad (2.13\text{a})$$

$$X(2)=X(1)\,[\tfrac{1}{2}-X(0)]\,, \qquad (2.13\text{b})$$

$$X(3)=X(1)\,[\tfrac{1}{6}-X(1)]\,,\quad \text{etc.} \qquad (2.13\text{c})$$

The relationships among the $Y(n)$'s are similar.

Following the procedure used in I, a series expansion for the correlations $\langle\sigma_g^\lambda\sigma_p^\nu\rangle$ may be calculated.[3] Such a series expansion for the relevant correlation has the form

$$\langle \sigma_g^A \sigma_p^C \rangle \equiv \{ m^A m^C [1 - \alpha^{AC}(\vec{g}\,\vec{p})] \}$$
$$= (1 - \delta_{gp}) \, [m^A m^C + \Phi_0^{AC}(\overrightarrow{gp}) + \beta \Phi_1^{AC}(\overrightarrow{gp})$$
$$+ \beta^2 \Phi_2^{AC}(\overrightarrow{gp}) + \beta^3 \Phi_3^{AC}(\overrightarrow{gp})] + O(1/T)^4 \, , \tag{2.14}$$

with the values of the $\Phi_i^{AC}(\overrightarrow{gp})$ given in Appendix A. Of course when A and C are interchanged with B in Eq. (2.14), we find the remaining two SRO parameters.

As for the corresponding case for the binary alloy,[2] it is convenient to use a series inversion for Eq. (2.14). However, to do this we proceed as in I, perform an inverse-lattice Fourier transformation and then, as in Ref. 2, carry out the series inversion in such a manner that all the K dependence occurs in the denominator.

In this manner we finally obtain

$$\alpha^{AC}(\vec{k}) = \frac{D(AC)}{1 + \beta \Phi_1^{AC}(\vec{k}) + \beta^2 \Phi_2^{AC}(\vec{k}) + \beta^3 \Phi_3^{AC}(\vec{k})} \, , \tag{2.15}$$

where the values of $\Phi_i^{AC}(\vec{k})$ are given in Appendix B.

In Eq. (2.15) the coefficient $D(AC)$ is determined such that the sum rule

$$\frac{1}{N} \sum_{\vec{k}} \alpha^{AC}(\vec{k}) = 1 \tag{2.16}$$

is exactly conserved. This step is central to our series-inversion procedure which was also followed in I and in Ref. 2.

Note that Eq. (2.15) contains singularities whereas the expansion (2.14) has no such character. This point has been fully discussed in Ref. 2 and, therefore, we shall not belabor it further. All that needs to be said here is that to the order of accuracy of Eq. (2.14), the two equations, i.e., Eqs. (2.14) and (2.15), yield identical high-temperature series expansions.

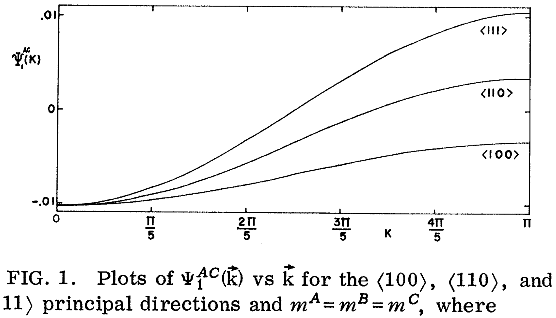

FIG. 1. Plots of $\Psi_1^{AC}(\vec{k})$ vs $\vec{k}$ for the ⟨100⟩, ⟨110⟩, and ⟨111⟩ principal directions and $m^A = m^B = m^C$, where $\gamma = 1.0$ and $\epsilon = 1.2$ ($\gamma = U^{BC}/U^{AC}$, $\epsilon = U^{AB}/U^{AC}$). Simple cubic nearest-neighbor interactions have been used.

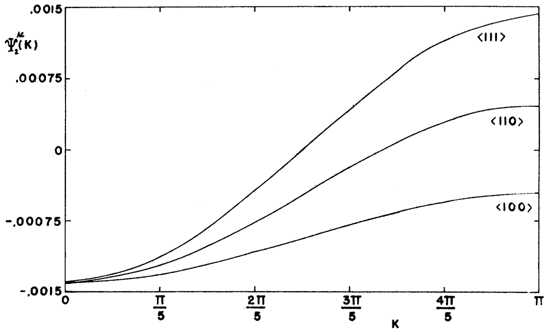

FIG. 2. Same as Fig. 1, except $\Psi_2^{AC}(\vec{k})$ vs $\vec{k}$.

III. DISCUSSIONS AND COMMENTS

The improved solutions for the SRO parameters given in Eq. (2.15) differ from the earlier linear approximation of I in that it contains the additional temperature-dependent terms involving $\Psi_2^{AC}(\vec{k})$ and $\Psi_3^{AC}(\vec{k})$, whose presence also renormalizes the coefficient $D(AC)$ through the sum rule given in (2.16). The limiting case of binary composition, i.e., where the relative concentration of one of the three components is vanishingly small, has already been studied in detail in Ref. 2. There it was noted that the presence of these terms caused a renormalization in the magnitudes of the SRO parameter and the transition temperature by several percent. The more important consequence of these terms, however, was that they led to a basically different dependence of the transition temperature upon the system composition and upon the range of interparticle potential than had been predicted by the linear theory. It is therefore reasonable to anticipate that the inclusion of the additional β^2 and β^3 terms in the expressions (2.15) and (2.16) will also affect significant improvements in the corresponding linear theory results for the ternary systems given in I.

To remark on the interesting question of how the improved solution affects the dependence of the transition temperature upon the interparticle potentials for those compositions where the concentra-

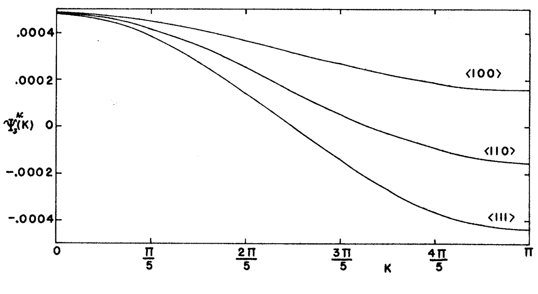

FIG. 3. Same as Fig. 1, except $\Psi_3^{AC}(\vec{k})$ vs $\vec{k}$.

TABLE I. Relative importance of the additional terms $\Psi_2^{AC}(\vec{k})$ and $\Psi_3^{AC}(\vec{k})$ with respect to the linear result $\Psi_1^{AC}(\vec{k})$ shown as a function of $\vec{k}$ and of the parameter $t = T_c/T$ $(k_B T_c = 6U^{AC})$. The cubic approximation result $G^{AC}(\beta, \vec{k})$ is defined via Eq. (3.1), e.g., $1 + tG^{AC}(t, \vec{k}) \equiv 1 + t\,[\Psi_1^{AC}(\vec{k}) + t\Psi_2^{AC}(\vec{k}) + t^2\Psi_3^{AC}(\vec{k})]$. Here we have taken the equicomposition case of $m^A = m^B = m^C = \frac{1}{3}$, with $\epsilon = U^{AB}/U^{AC} = 1.2$, $\gamma = U^{BC}/U^{AC} = 1.0$, and the principal direction is $\langle 100 \rangle$.

t	$\vec{k}$	$\Psi_1^{AC}(\vec{k})$	$G^{AC}(t, \vec{k})$	$t\Psi_2^{AC}(\vec{k})$	$t^2\Psi_3^{AC}(\vec{k})$
0.95	0	-0.103704	-0.104605	-0.001338	0.000437
0.95	$\frac{1}{2}\pi$	-0.069126	-0.069742	-0.000892	0.000286
0.95	π	-0.034568	-0.034873	-0.000446	0.000141
0.75	0	-0.103704	-0.104488	-0.001056	0.000272
0.75	$\frac{1}{2}\pi$	-0.069136	-0.069662	-0.000704	0.000178
0.75	π	-0.034568	-0.034832	-0.000352	0.000088
0.50	0	-0.103704	-0.104287	-0.000704	0.000121
0.50	$\frac{1}{2}\pi$	-0.069136	-0.069526	-0.000470	0.000080
0.50	π	-0.034568	-0.034764	-0.000235	0.000039
0.25	0	-0.103704	-0.104026	-0.000352	0.000030
0.25	$\frac{1}{2}\pi$	-0.069136	-0.069351	-0.000235	0.000020
0.25	π	-0.034568	-0.034676	-0.000117	0.000010

TABLE II. Same as Table I except the principal direction is $\langle 110 \rangle$.

t	$\vec{k}$	$\Psi_1^{AC}(\vec{k})$	$G^{AC}(t, \vec{k})$	$t\Psi_2^{AC}(\vec{k})$	$t^2\Psi_3^{AC}(\vec{k})$
0.95	0	-0.103704	-0.104605	-0.001338	0.000437
0.95	$\frac{1}{2}\pi$	-0.034568	-0.034873	-0.000446	0.000141
0.95	π	0.034568	0.034877	0.000446	-0.000136
0.75	0	-0.103704	-0.104488	-0.001056	0.000272
0.75	$\frac{1}{2}\pi$	-0.034568	-0.034832	-0.000352	0.000088
0.75	π	0.034568	0.034835	0.000352	-0.000085
0.50	0	-0.103704	-0.104287	-0.000704	0.000121
0.50	$\frac{1}{2}\pi$	-0.034568	-0.034764	-0.000235	0.000039
0.50	π	0.034568	0.034765	0.000235	-0.000038
0.25	0	-0.103704	-0.104026	-0.000352	0.000030
0.25	$\frac{1}{2}\pi$	-0.034568	-0.034676	-0.000117	0.000010
0.25	π	0.034568	0.034676	0.000117	-0.000009

TABLE III. Same as Table I except the principal direction is $\langle 111 \rangle$.

t	$\vec{k}$	$\Psi_1^{AC}(\vec{k})$	$G^{AC}(t, \vec{k})$	$t\Psi_2^{AC}(\vec{k})$	$t^2\Psi_3^{AC}(\vec{k})$
0.95	0	-0.103704	-0.104605	-0.001338	0.000437
0.95	$\frac{1}{2}\pi$	0.000000	0.000000	0.000000	0.000000
0.95	π	0.103704	0.104646	0.001338	-0.000395
0.75	0	-0.103704	-0.104488	-0.001056	0.000272
0.75	$\frac{1}{2}\pi$	0.000000	0.000000	0.000000	0.000000
0.75	π	0.103704	0.104514	0.001056	-0.000246
0.50	0	-0.103704	-0.104287	-0.000704	0.000121
0.50	$\frac{1}{2}\pi$	0.000000	0.000000	0.000000	0.000000
0.50	π	0.103704	0.104299	0.000704	-0.000110
0.25	0	-0.103704	-0.104026	-0.000352	0.000030
0.25	$\frac{1}{2}\pi$	0.000000	0.000000	0.000000	0.000000
0.25	π	0.103704	0.104029	0.000352	-0.000027

tion of one of the components is close to 50%, we first note that for such a case the linear approximation of I leads to a particularly unphysical prediction, namely, that for the case when m^A is $\frac{1}{2}$ and the interparticle potentials are such that $(k_B T_c)^{AC}$ is the highest of the three[4] temperatures $(k_B T_c)^{\lambda\nu}$, then the transition temperature is entirely independent of the strength of the interparticle potential U^{AB}. In the improved approximation of the present paper, this unphysical feature of the linear approximation is removed and a nontrivial dependence of the transition temperature $(k_B T_c)^{AC}$ on U^{AB} is found.

However, in contrast to the behavior of the critical temperature, the predictions of the present theory regarding the behavior of the SRO parameters and their inverse Fourier transforms, e.g., $\alpha^{AC}(\vec{k})$, are not substantially different from those of the linear case. It is, nevertheless, interesting to examine the relative importance of the additional terms which have been introduced into Eqs. (2.15) and (2.16). Such a comparison would bear upon the convergence properties of the series-inversion procedure used here.

To get a feel for the variation of the temperature-independent parameters $\Psi_1^{AC}(\vec{k})$, $\Psi_2^{AC}(\vec{k})$, and $\Psi_3^{AC}(\vec{k})$, these parameters have been plotted in Fig. 1–3. These curves have been plotted for the equiconcentration case $m^A = m^B = m^C$ and assuming $U^{AC} = U^{BC} = \frac{5}{6}U^{AB}$. Note that the dominant term $\Psi_1^{AC}(\vec{k})$, has a minimum at $\vec{k} = (0, 0, 0)$ where the SRO parameter $\alpha^{AC}(\vec{k})$ achieves its maximum value. Also if $\Psi_2^{AC}(\vec{k})$ and $\Psi_3^{AC}(\vec{k})$ are assumed to be equal to zero, one retrieves the linear-approximation results of I. Similar plots can also be presented for $\alpha^{AB}(\vec{k})$ and $\alpha^{BC}(\vec{k})$, but for brevity we do not include them here.

For a convenient comparison of the magnitudes of the two new terms, involving $\Psi_2^{AC}(\vec{k})$ and $\Psi_3^{AC}(\vec{k})$, in the denominator of Eq. (2.15) with the old term

TABLE IV. Values of the SRO parameters $\alpha^{\mu\nu}(\vec{k})$, $\mu, \eta = A, B, C$, for the linear (L) and cubic (c) cases. The equicomposition case, $m^A = m^B = m^C = \frac{1}{3}$, has been used, $\epsilon = U^{AB}/U^{AC} = 1.2$, $\gamma = U^{BC}/U^{AC} = 1.0$, and $T_c/T = \frac{2}{3}$ $(k_B T_c = 6U^{AC})$. Computations are along the $\langle 100 \rangle$ principal direction for simple cubic nearest-neighbor interactions.

$\vec{k}$	$\alpha_L^{AC}(\vec{k})$	$\alpha_c^{AC}(\vec{k})$	$\alpha_L^{BC}(\vec{k})$	$\alpha_c^{BC}(\vec{k})$	$\alpha_L^{AB}(\vec{k})$	$\alpha_c^{AB}(\vec{k})$
0	1.0734	1.0740	1.0734	1.0740	1.1078	1.1077
$\frac{1}{10}\pi$	1.0721	1.0727	1.0721	1.0727	1.1058	1.1057
$\frac{2}{10}\pi$	1.0684	1.0689	1.0684	1.0689	1.1001	1.1001
$\frac{3}{10}\pi$	1.0626	1.0630	1.0626	1.0630	1.0913	1.0913
$\frac{4}{10}\pi$	1.0554	1.0558	1.0554	1.0558	1.0805	1.0805
$\frac{5}{10}\pi$	1.0475	1.0478	1.0475	1.0478	1.0687	1.0688
$\frac{6}{10}\pi$	1.0397	1.0400	1.0397	1.0400	1.0572	1.0573
$\frac{7}{10}\pi$	1.0328	1.0330	1.0328	1.0330	1.0471	1.0471
$\frac{8}{10}\pi$	1.0274	1.0276	1.0274	1.0276	1.0391	1.0392
$\frac{9}{10}\pi$	1.0240	1.0241	1.0240	1.0241	1.0341	1.0342
π	1.0228	1.0229	1.0228	1.0229	1.0324	1.0324

TABLE V. Same as Table IV except $\gamma = 1.5$.

$\vec{k}$	$\alpha_L^{AC}(\vec{k})$	$\alpha_c^{AC}(\vec{k})$	$\alpha_L^{BC}(\vec{k})$	$\alpha_c^{BC}(\vec{k})$	$\alpha_L^{AB}(\vec{k})$	$\alpha_c^{AB}(\vec{k})$
0	1.0596	1.0583	1.1473	1.1454	1.0932	1.0928
$\frac{1}{10}\pi$	1.0586	1.0574	1.1445	1.1427	1.0915	1.0911
$\frac{2}{10}\pi$	1.0556	1.0545	1.1364	1.1349	1.0867	1.0863
$\frac{3}{10}\pi$	1.0509	1.0502	1.1240	1.1228	1.0792	1.0789
$\frac{4}{10}\pi$	1.0451	1.0447	1.1088	1.1080	1.0699	1.0697
$\frac{5}{10}\pi$	1.0388	1.0386	1.0924	1.0920	1.0598	1.0597
$\frac{6}{10}\pi$	1.0325	1.0325	1.0765	1.0763	1.0499	1.0499
$\frac{7}{10}\pi$	1.0269	1.0270	1.0626	1.0626	1.0411	1.0411
$\frac{8}{10}\pi$	1.0225	1.0227	1.0518	1.0519	1.0342	1.0343
$\frac{9}{10}\pi$	1.0197	1.0200	1.0449	1.0451	1.0299	1.0299
π	1.0187	1.0190	1.0426	1.0428	1.0284	1.0285

TABLE VI. Same as Table IV except the principal direction is $\langle 111 \rangle$.

$\vec{k}$	$\alpha_L^{AC}(\vec{k})$	$\alpha_c^{AC}(\vec{k})$	$\alpha_L^{BC}(\vec{k})$	$\alpha_c^{BC}(\vec{k})$	$\alpha_L^{AB}(\vec{k})$	$\alpha_c^{AB}(\vec{k})$
0	1.0734	1.0740	1.0734	1.0740	1.1078	1.107
$\frac{1}{10}\pi$	1.0695	1.0700	1.0695	1.0700	1.1019	1.1018
$\frac{2}{10}\pi$	1.0584	1.0588	1.0584	1.0588	1.0851	1.0851
$\frac{3}{10}\pi$	1.0415	1.0418	1.0415	1.0418	1.0599	1.0600
$\frac{4}{10}\pi$	1.0210	1.0212	1.0210	1.0212	1.0298	1.0299
$\frac{5}{10}\pi$	0.9992	0.9992	0.9992	0.9992	0.9984	0.9984
$\frac{6}{10}\pi$	0.9783	0.9781	0.9783	0.9781	0.9688	0.9688
$\frac{7}{10}\pi$	0.9602	0.9599	0.9602	0.9599	0.9436	0.9434
$\frac{8}{10}\pi$	0.9463	0.9459	0.0463	0.9459	0.9134	0.9242
$\frac{9}{10}\pi$	0.9376	0.9371	0.9376	0.9371	0.9126	0.9123
π	0.9346	0.9341	0.9346	0.9341	0.9086	0.9083

involving only $\Psi_1^{AC}(\vec{k})$ that was obtained within the linear approximation, we have included Tables I–III. These tables give a description of the relative importance of the new and the old terms as a function of the system temperature. Here we have used the notation,

$$1 + \beta\Psi_1^{AC}(\vec{k}) + \beta^2\Psi_2^{AC}(\vec{k}) + \beta^3\Psi_3^{AC}(\vec{k}) \equiv 1 + \beta G^{AC}(\beta, \vec{k}) \ . \tag{3.1}$$

Of course as the system temperature becomes large, the new terms become increasingly unimportant and then the results of the linear theory become accurate.

It should be emphasized here that while a study of Tables I–III convinces one of the rapid convergence of the high-temperature series expansions, the effects of the inclusion of the additional terms involving $\Psi_2^{AC}(\vec{k})$ and $\Psi_3^{AC}(\vec{k})$ in Eq. (2.15) are nontrivial. The most important consequence of these additional terms, of course, is a change in the position of the singularity, which results in the changed behavior of the transition temperature as a function of the interparticle potentials and the system concentration. The less important consequence is a change in the magnitude of the SRO parameters as compared with the corresponding results of the linear approximation (see Tables IV–VI).

It should be noted that although, for convenience, the numerical results of this paper have only been worked out for the simple case of separating ternaries with only nearest-neighbor interactions, the formal analysis presented here is equally applicable to ordering alloys as well as to the case of larger-range potentials. Indeed, it is hoped that in a future publication a detailed computation of the transition temperature and SRO parameters, making use of relatively realistic potentials and compositions, will be given.

The conclusions of this paper may be briefly recapitulated by saying (a) that while the linear approximation of I can be expected to be valid only at elevated temperatures where $T \gg T_c$, the expressions for the SRO parameters presented here should be adequate even in the region of intermediate temperatures $T \gtrsim T_c$ and (b) moreover, the obviously unphysical feature of the linear approximation, regarding the dependence of the transition temperature upon the system composition when one of the concentrations is close to 50%, is eliminated in this improved approximation.

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APPENDIX A

The expressions for $\Phi_0^{AC}(\vec{g}\,\vec{p})$, $\Phi_1^{AC}(\vec{g}\,\vec{p})$, $\Phi_2^{AC}(\vec{g}\,\vec{p})$, and $\Phi_3^{AC}(\vec{g}\,\vec{p})$ in Eq. (2.14) are given as follows (the corresponding expressions for $\alpha^{AB}(\vec{g}\,\vec{p})$ and $\alpha^{BC}(\vec{g}\,\vec{p})$ are found by interchanging $A \leftrightarrow B \leftrightarrow C$; note that W(gp) also undergoes a change):

$$\Phi_0^{AC}(\vec{g}\,\vec{p}) \equiv -\,\delta_{gp}\Big\{ m^A m^C + \beta^2 m^A (m^C)^2 \sum_f \,[m^A W(gf) - (1 - m^C)\, 2U^{BC}(gf)]\,\Gamma^{AC}(gf) + \beta^2 (m^A)^2 m^C$$

$$\times \sum_f \, [2m^A U^{AB}(gf) - (1-m^C)\, W(gf)] \;\; \Gamma^{AA}(gf)$$

$$+\beta^3 \sum_f \Big[[(m^A)^2 m^C/(1-m^C)] \Big(m^A m^C [2m^A U^{AB}(gf) - (1-m^C)\, W(gf)] \{\Gamma^{AA}(gf)$$

$$\times\; [2U^{AB}(gf)\,(1-2m^A) - m^C W(gf)] - m^C W(gf)\, \Gamma^{AC}(gf)\} - (m^C)^2\, [2U^{BC}(gf)\,(1-m^C) - m^A W(gf)]$$

$$\times \{\Gamma^{AC}(gf)\, [W(gf)\,(1-2m^C) - 2m^A U^{AB}(gf)] - 2m^A U^{AB}(gf)\, \Gamma^{AA}(gf)\}$$

$$-\,\Gamma^{AC}(gf)\, \{\Gamma^{AC}(gf)\, [m^C(1-2m^A)\, 2U^{AB}(gf) - (m^C)^2 W(gf)] - (m^C)^2 W(gf)\, \Gamma^{CC}(gf)\}$$

$$+\tfrac{1}{2}(m^B - m^A)\, \big(m^A(1-2m^A)\, [2\, U^{AB}(gf)]^2\, \Gamma^{AA}(gf) + m^C(1-2m^C)\, W(gf)^2\, \Gamma^{AC}(gf)$$

$$-\, 2m^A m^C\, W(gf)\, 2U^{AB}(gf)\, [\Gamma^{AA}(gf) + \Gamma^{AC}(gf)] - \Gamma^{AC}(gf)\, \{(1-m^A)(1-2m^A)$$

$$\times [2U^{AB}(gf)]^2 - m^C(1-2m^C) W(gf)^2 - 2m^C(1-2m^A) W(gf) 2U^{AB}(gf)\}\big)\Big) - [m^A m^C(1-m^C)/(1-m^A)]\big((m^A)^2$$

$$\times [2U^{AB}(gf)(1-m^A) - m^C W(gf)]\{\, \Gamma^{AA}(gf)\, [m^C 2U^{BC}(gf) - (1-2m^A) W(gf)] + m^C 2U^{BC}(gf)\Gamma^{AC}(gf)\}$$

$$+ m^A m^C [2m^C U^{BC}(gf) - (1-m^A) W(gf)]\, \{\, \Gamma^{AC}(gf)[(1-2m^C) 2U^{BC}(gf) - m^A W(gf)] - m^A W(gf) \Gamma^{AA}(gf)\}$$

$$-\tfrac{1}{2}(m^C - m^B)\, \{m^A(1-2m^A) W(gf)^2 \Gamma^{AA}(gf) + m^C(1-2m^C)[2U^{BC}(gf)]^2 \Gamma^{AC}(gf) - 2m^A m^C\, W(gf) 2U^{BC}(gf)$$

$$\times\;\; [\Gamma^{AA}(gf) + \Gamma^{AC}(gf)]\}\big) - [m^A (m^C)^2/(1-m^A)]\, \Gamma^{CC}(gf)\big(\, \Gamma^{AA}(gf)[W(gf) m^A (1-2m^A) - 2m^A m^C U^{BC}(gf)]$$

$$-\, 2m^A m^C U^{BC}(gf) \Gamma^{AC}(gf) - \tfrac{1}{2}(m^C - m^B)\{(1-m^A)(1-2m^A) W(gf)^2 - m^C(1-2m^C)[2U^{BC}(gf)]^2$$

$$-\, 2m^C(1-2m^A) W(gf) 2U^{BC}(gf)\}\big) \Big] + \beta^3 \sum_{f_1 f_2} \; \{(m^A)^3 m^C \Gamma^{AA}(gf_2)[W(gf_1)(1-m^C) - 2m^A U^{AB}(gf_1)][m^C W(f_1 f_2)$$

$$-\,(1-m^A) 2U^{AB}(f_1 f_2)] + (m^A m^C)^2 \Gamma^{AC} \;\, (gf_2)[W(gf_1)(1-m^C) - 2m^A U^{AB}(gf_1)][2m^C U^{BC}(f_1 f_2) - (1-m^A) W(f_1 f_2)]$$

$$+\,(m^A m^C)^2 \Gamma^{AA}(gf_2)[W(gf_1) m^A - (1-m^C) 2U^{BC}(gf_1)][W(f_1 f_2)(1-m^C) - 2m^A U^{AB}(f_1 f_2)]$$

$$+\, m^A (m^C)^3 \Gamma^{AC}(gf_2)[m^A W(gf_1) - (1-m^C) 2U^{BC}(gf_1)][(1-m^C) 2U^{BC}(f_1 f_2) - m^A W(f_1 f_2)]\} \Big\} \;\;, \qquad \text{(A1)}$$

$$\Phi_1^{AC}(\vec{g}\vec{p}) \equiv -\, m^A m^C \Gamma^{AC}(gp)\;, \qquad \text{(A2)}$$

$$\Phi_2^{AC}(\vec{g}\vec{p}) \equiv [m^A m^C (1-m^C)/(1-m^A)]\big(\Gamma^{AA}(gp)[m^A(1-2m^A) W(gp) - 2m^A m^C U^{BC}(gp)] - 2m^A m^C U^{BC}(gp) \Gamma^{AC}(gp)$$

$$-\tfrac{1}{2}(m^C - m^B)\, \{(1-m^A)(1-2m^A) W(gp)^2 - m^C(1-2m^C)[2U^{BC}(gp)]^2 - 2m^C(1-2m^A) W(gp) 2\, U^{BC}(gp)\}\big)$$

$$-\, [(m^A)^2 m^C/(1-m^C)]\big([m^C(1-2m^A)\;\, 2U^{AB}(gp) - (m^C)^2 W(gp)]\Gamma^{AC}(gp) - (m^C)^2 W(gp) \Gamma^{CC}(gp)$$

$$+\tfrac{1}{2}\,(m^B - m^A)\, \{(1-m^A)\,(1-2m^A)\,[2U^{AB}(gp)]^2 - m^C(1-2m^C)\; W(gp)^2 - 2m^C(1-2m^A)\; W(gp)\; 2U^{AB}(gp)\}\big)$$

$$+\, m^A\, m^C \textstyle\sum_f \{m^A\, \Gamma^{AA}(pf)\, [2m^A\, U^{AB}(gf) - (1-m^C)\, W(gf)] \;\; + m^C\, \Gamma^{AC}(pf)\, [m^A\, W(gf) - (1-m^C)\, 2U^{BC}(gf)]\}\;, \quad \text{(A3)}$$

$$\Phi_3^{AC}(\vec{g}\,\vec{p}) \equiv \sum_f \Big\{ [(m^A)^2 m^C/(1-m^C)] \Big[m^A m^C [2U^{AB}(gf)\, m^A - (1-m^C)\; W(gf)]\, \{\Gamma^{AA}(pf)\, [(1-2m^A)\, 2U^{AB}(gf) - m^C\; W(gf)]$$

$$-\, m^C\, W(gf)\;\; \Gamma^{AC}(pf)\} - (m^C)^2\, [2U^{BC}(gf)(1-m^C) - m^A\; W(gf)]\, \{\Gamma^{AC}(pf)\, [W(gf)(1-2m^C) \;- m^A\; 2U^{AB}(gf)]$$

$$-\, m^A\; 2U^{AB}(gf)\; \Gamma^{AA}(pf)\} - m^C\;\, \Gamma^{AC}(pf)\, \{\Gamma^{AC}(gf)\, [2U^{AB}(gf)(1-2m^A) - m^C\;\, W(gf)] - m^C\;\, W(gf)\; \Gamma^{CC}(gf)\}$$

$$+\tfrac{1}{2}\,(m^B - m^A)\, \big(m^A(1-2m^A)\, [2U^{AB}(gf)]^2\, \Gamma^{AA}(pf) + m^C(1-2m^C)\, W(gf)^2\, \Gamma^{AC}(pf) - 2m^A\, m^C\;\, W(gf)\; 2U^{AB}(gf)$$

$$\times [\Gamma^{AA}(pf) + \Gamma^{AC}(pf)] - \Gamma^{AC}(pf)\, \{(1-m^A)(1-2m^A)\; [2U^{AB}(gf)]^2 - m^C(1-2m^C)\; W(gf)^2$$

$$-2m^C(1-2m^A)\,W(gf)\,2U^{AB}(gf)\}\big)\Big]-[m^A m^C/(1-m^A)]\Big[(m^A)^2(1-m^C)\,[2U^{AB}(gf)(1-m^A)-m^C\,W(gf)]$$

$$\times\;\{\Gamma^{AA}(pf)\,[2m^C\,U^{BC}(gf)\;-(1-2m^A)\,W(gf)]+2m^C\,U^{BC}(gf)\,\Gamma^{AC}(pf)\}+m^Am^C(1-m^C)$$

$$\times[2m^C\,U^{BC}(gf)-(1-m^A)\,W(gf)]\{\Gamma^{AC}(pf)\,[2U^{BC}(gf)(1-2m^C)-m^A\,W(gf)]-m^A\,W(gf)\,\Gamma^{AA}(pf)\}+m^C\,\Gamma^{CC}(pf)$$

$$\times\;\{\Gamma^{AA}(gf)\,[m^A(1-2m^A)\,W(gf)-2m^Am^C\,U^{BC}(gf)]-2m^Am^C\,U^{BC}(gf)\,\Gamma^{AC}(gf)\}$$

$$-\tfrac{1}{2}\,(m^C-m^B)\,\big(m^A(1-m^C)(1-2m^A)\,W(gf)^2\,\Gamma^{AA}(pf)+m^C(1-m^C)\,(1-2m^C)\,[2U^{BC}(gf)]^2\,\Gamma^{AC}(pf)$$

$$-2m^Am^C(1-m^C)\,W(gf)\;\;2U^{BC}(gf)\,[\Gamma^{AA}(pf)+\Gamma^{AC}(pf)]+m^C\,\Gamma^{CC}(pf)\,\{(1-m^A)$$

$$\times(1-2m^A)\,W(gf)^2-m^C(1-2m^C)\,[2U^{BC}(gf)]^2-2m^C\,(1-2m^A)\,W(gf)\,2U^{BC}(gf)\}\big)\Big]\Big\}$$

$$+\sum_f\Big[\;(m^A)^2\,m^C\;[W(gp)(1-m^C)-2m^A\,U^{AB}(gp)]\,\{m^A\,\Gamma^{AA}(gf)\,[2U^{AB}(gf)(1-m^A)-m^C\,W(gf)]$$

$$+m^C\,\Gamma^{AC}(gf)\,[W(gf)(1-m^A)-2m^C\,U^{BC}(gf)]\}\;+m^A(m^C)^2\,[2U^{BC}(gp)(1-m^C)-m^A\,W(gp)]\,\{m^A\,\Gamma^{AA}(gf)$$

$$\times[(1-m^C)\,W(gf)-2m^A\,U^{AB}(gf)]+m^C\,\Gamma^{AC}(gf)\,[(1-m^C)\;2U^{BC}(gf)-m^A\,W(gf)]\}$$

$$+\frac{m^Am^C\,(1-m^C)[(1-m^A)^2-6m^Bm^C]}{2(1-m^A)^2}\,\big([(1-m^A)\,W(gp)-2m^C\,U^{BC}(gp)]\,\{m^A(1-m^A)\,W(gf)^2+m^C(1-m^C)$$

$$\times[2U^{BC}(gf)]^2-2m^Am^C\,W(gf)\,2U^{BC}(gf)\}\big)-[m^C(m^A)^2/2(1-m^C)^2]\,[(1-m^C)^2-6m^Am^B]$$

$$\times\;\big([(1-m^A)\,2U^{AB}(gp)-m^C\,W(gp)]\;\{m^A(1-m^A)\,[2U^{AB}(gf)]^2+m^C(1-m^C)\,W(gf)^2-2m^Am^C$$

$$\times W(gf)\,2U^{AB}(gf)\}\big)-\frac{m^Am^C(1-m^C)(m^C-m^B)}{2(1-m^A)^2}\,\{m^A(1-m^A)\,W(gf)^2+m^C(1-m^C)\,[2U^{BC}(gf)]^2$$

$$-2m^Am^C\,W(gf)\,2U^{BC}(gf)\}\;\{m^A\,\Gamma^{AA}(gp)-(m^C-m^B)\,[2m^C\,U^{BC}(gp)-(1-m^A)\,W(gp)]\}$$

$$-\frac{m^C(m^A)^2(m^B-m^A)}{2(1-m^C)^2}\,\{m^A(1-m^A)\,[2U^{AB}(gf)]^2+m^C(1-m^C)\,W(gf)^2-2m^Am^C\,W(gf)\,2U^{AB}(gf)\}$$

$$\times\;\;\{m^C\Gamma^{AC}(gp)+(m^B-m^A)\,[m^C\,W(gp)-(1-m^A)\,2U^{AB}(gp)]\}-\frac{m^C\,(m^A)^2(1-m^C)}{(1-m^A)^2}\,\Gamma^{AA}(gp)$$

$$\times\big((m^A)^2\,\Gamma^{AA}(gf)\,W(gf)+2m^Am^C\,\Gamma^{AC}(gf)\,U^{BC}(gf)-\tfrac{1}{2}(m^C-m^B)\,\{m^A(1-m^A)\,W(gf)^2+m^C\,(1-m^C)\,[2U^{BC}(gf)]^2$$

$$-2m^Am^C\;W(gf)\,2U^{BC}(gf)\}\big)+[m^Am^C/(1-m^C)]^2\,\Gamma^{AC}(gp)\,\big(m^Am^C\,\Gamma^{AC}(gf)$$

$$\times 2U^{AB}(gf)+(m^C)^2W(gf)\Gamma^{CC}(gf)+\tfrac{1}{2}\,(m^B-m^A)\{m^A(1-m^A)\;[2U^{AB}\,(gf)]^2+m^C(1-m^C)W(gf)^2$$

$$-2m^Am^C\,W(gf)2U^{AB}(gf)\}\big)+[m^Am^C/(1-m^A)]^2\,\big(m^A[(1-m^A)2U^{AB}(gf)+(1-m^C)W(gf)]$$

$$\times\{\Gamma^{AA}(gf)[m^A(1-m^A)W(gp)-2m^Am^C\,U^{BC}(gp)]+\Gamma^{AA}(gp)\;[m^A(1-m^A)W(gf)-2m^Am^C\,U^{BC}(gf)]\}$$

$$-m^C[(1-m^A)W(gf)+2(1-m^C)U^{BC}(gf)]\{\Gamma^{AA}(gp)[(m^A)^2W(gf)-m^A(1-m^C)2U^{BC}(gf)]$$

$$-\Gamma^{AC}(gf)[\,W(gp)m^A(1-m^A)-2m^Am^C\,U^{BC}(gp)]\}-(m^C-m^B)\;\{[m^A(1-m^A)2U^{AB}(gf)+m^A(1-m^C)W(gf)]$$

$$\times[(1-m^A)W(gf)-2m^C\,U^{BC}(gf)]-m^C[(1-m^A)W(gf)+(1-m^C)2U^{BC}(gf)]$$

$$\times[m^A W(gf)-(1-m^C)2U^{BC}(gf)]\}[(1-m^A)W(gp)-2m^C U^{BC}(gp)]\Big)-[(m^A)^2 m^C/(1-m^C)^2]$$

$$\times\Big(m^A[(1-m^C)^2 W(gf)+2m^A m^C U^{AB}(gf)]\{\Gamma^{AC}(gf)[m^C(1-m^A)2U^{AB}(gp)-(m^C)^2 W(gp)]+\Gamma^{AC}(gp)$$

$$\times[m^C(1-m^A)2U^{AB}(gf)-(m^C)^2 W(gf)]\}-m^C[(1-m^C)^2 2U^{BC}(gf)+m^A m^C W(gf)]$$

$$\times\ \{\Gamma^{AC}(gp)[2m^A m^C U^{AB}(gf)-m^C(1-m^C)W(gf)]-\Gamma^{CC}(gf)[m^C(1-m^A)2U^{AB}(gp)-(m^C)^2 W(gp)]\}$$

$$+(m^B-m^A)[(1-m^A)2U^{AB}(gp)-m^C W(gp)]$$

$$\times\ \{[(1-m^C)^2 W(gf)+2m^A m^C U^{AB}(gf)][m^A(1-m^A)2U^{AB}(gf)-m^A m^C W(gf)]-m^C[(1-m^C)^2 2U^{BC}(gf)$$

$$+m^A m^C W(gf)][2m^A m^C U^{AB}(gf)-m^C(1-m^C)W(gf)]]\}\Big)\Bigg]+\sum_{f_1 f_2}\{(m^A)^3 m^C\Gamma^{AA}(pf_2)$$

$$\times\ [(1-m^C)W(gf_1)-2m^A U^{AB}(gf_1)]\ \ [m^C W(f_1 f_2)-(1-m^A)2U^{AB}(f_1 f_2)]+(m^A m^C)^2\Gamma^{AA}(pf_2)$$

$$\times[m^A W(gf_1)-(1-m^C)2U^{BC}(gf_1)][(1-m^C)W(f_1 f_2)-2m^A U^{AB}(f_1 f_2)$$

$$+(m^A m^C)^2\Gamma^{AC}(pf_2)[(1-m^C)W(gf_1)-2m^A U^{AB}(gf_1)][2m^C U^{BC}(f_1 f_2)-(1-m^A)W(f_1 f_2)]$$

$$+m^A(m^C)^3\Gamma^{AC}(pf_2)[m^A W(gf_1)-(1-m^C)2U^{BC}(gf_1)][(1-m^C)\ 2U^{BC}(f_1 f_2)-m^A W(f_1 f_2)]\}$$

$$+\sum_f\Bigg[[m^A m^C/(1-m^C)]\Big(m^A m^C(1-2m^A)[2m^A U^{AB}(gf)\Gamma^{AA}(pf)+m^C W(gf)\Gamma^{AC}(pf)]$$

$$\times\ [2m^A U^{AB}(gp)-(1-m^C)\ W(gp)]+m^A(m^C)^2\{2m^A U^{AB}(gf)[\Gamma^{AA}(pf)+\Gamma^{AC}(pf)]$$

$$+m^C W(gf)[\Gamma^{AC}(pf)+\Gamma^{CC}(pf)]\}[2U^{BC}(gp)(1-m^C)-m^A W(gp)]+m^A(m^C)^2\{(1-2m^A)2U^{AB}(gp)\Gamma^{AC}(pf)$$

$$-m^C W(gp)[\Gamma^{AC}(pf)+\Gamma^{CC}(pf)]\}[m^A W(gf)-(1-m^C)2U^{BC}(gf)]+m^C(m^A)^2$$

$$\times\{(1-2m^A)2U^{AB}(gp)\Gamma^{AA}(pf)-m^C W(gp)[\Gamma^{AA}(pf)+\Gamma^{AC}(pf)]\}\ [2m^A U^{AB}(gf)-(1-m^C)W(gf)]$$

$$+(m^B-m^A)\{(m^A)^2\ (1-2m^A)2U^{AB}(gp)2U^{AB}(gf)\Gamma^{AA}(pf)-m^A(m^C)^2 W(gp)W(gf)\ [\Gamma^{AC}(pf)+\Gamma^{CC}(pf)]$$

$$-m^C(m^A)^2 W(gp)2U^{AB}(gf)[\Gamma^{AA}(pf)+\Gamma^{AC}(pf)]\ +m^A m^C(1-2m^A)2U^{AB}(gp)W(gf)\Gamma^{AC}(pf)\}\Big)$$

$$-[m^C(1-m^C)/(1-m^A)]\ \Big((m^A)^2(1-2m^A)[m^A W(gf)\Gamma^{AA}(pf)+2m^C U^{BC}(gf)\Gamma^{AC}(pf)]$$

$$\times[m^C W(gp)-(1-m^A)2U^{AB}(gp)]+(m^A)^2 m^C\{m^A W(gf)$$

$$\times\ [\Gamma^{AA}(pf)+\Gamma^{AC}(pf)]+2m^C U^{BC}(gf)\ [\Gamma^{AC}(pf)+\Gamma^{CC}(pf)]\ \}$$

$$\times[(1-m^A)\ W(gp)-2m^C U^{BC}(gp)]+(m^A)^3\ [m^C W(gf)-(1-m^A)\ 2U^{AB}(gf)]\ \{W(gp)\ \Gamma^{AA}(pf)(1-2m^A)$$

$$-2m^C U^{BC}(gp)\ [\Gamma^{AA}(pf)+\Gamma^{AC}(pf)]\ \}+(m^A)^2 m^C\ \{W(gp)\Gamma^{AC}(pf)(1-2m^A)-2m^C U^{BC}(gp)$$

$$\times[\Gamma^{AC}(pf)+\Gamma^{CC}(pf)]\}[2m^C U^{BC}(gf)-(1-m^A)W(gf)]-(m^C-m^B)\ \{(m^A)^2(1-2m^A)W(gp)W(gf)\Gamma^{AA}(pf)$$

$$-m^A(m^C)^2 2U^{BC}(gp)\,2U^{BC}(gf)\ [\Gamma^{AC}(pf)+\Gamma^{CC}(pf)]\ -m^C(m^A)^2 2U^{BC}(gp)W(gf)[\Gamma^{AA}(pf)$$

$$+\Gamma^{AC}(pf)]\ +m^A m^C(1-2m^A)W(gp)2U^{BC}(gf)\Gamma^{AC}(pf)\}\Big)\Bigg]+\Bigg\{\frac{(m^A)^2 m^C[(1-m^C)^2-6m^A m^B]}{6(1-m^C)^2}$$

$$\times\ \{(1-m^A)[6m^A(1-m^A)-1]\ [2U^{AB}(gp)]^3+3W(gp)^2\ 2U^{AB}(gp)m^C(6m^A m^C+2m^B-1)$$

$$-3W(gp)[2U^{AB}(gp)]^2 m^C[6m^A(1-m^A)-1]-W(gp)^3 m^C\ \ [6m^C(1-m^C)-1]\ \}$$

$$
\begin{aligned}
&-\frac{m^A m^C(1-m^C)}{6(1-m^A)^2}\,[(1-m^A)^2-6m^B m^C]\,\{W(gp)^3(1-m^A)[6m^A(1-m^A)-1]+3W(gp)[2U^{BC}(gp)]^2\\
&\times m^C(6m^A m^C+2m^B-1)-3W(gp)^2\,2U^{BC}(gp)m^C[6m^A(1-m^A)-1]-[2U^{BC}(gp)]^3m^C\,[6m^C(1-m^C)-1]\}\\
&-\frac{(m^A)^2m^C(1-m^C)}{(1-m^A)^2}\,\Big[m^A m^C\,\Gamma^{AA}(gp)\,\{W(gp)^2[6m^A(1-m^A)-1]+2m^C(1-4m^A)\\
&\times W(gp)2U^{BC}(gp)+m^C(1-2m^C)[2U^{BC}(gp)]^2\}+m^A m^C\,\Gamma^{AC}(gp)\,\{2m^C(1-2m^A)W(gp)2U^{BC}(gp)\\
&+m^C(1-4m^C)[2U^{BC}(gp)]^2\}-\tfrac{1}{2}(m^C-m^B)(\Gamma^{AA}(gp)\,\{W(gp)^2\,[6m^A(1-m^A)-1]\;+m^C(1-2m^C)\,[2U^{BC}(gp)]^2\\
&+2m^C(1-4m^A)W(gp)2U^{BC}(gp)\}\;+\Gamma^{AC}(gp)\,\{m^C(1-4m^C)[2U^{BC}(gp)]^2+2m^C(1-2m^A)W(gp)2U^{BC}(gp)\}\\
&+W(gp)^3m^C(1-m^A)[6m^A(1-m^A)-1]-[2U^{BC}(gp)]^3(m^C)^2\,[6m^C(1-m^C)-1]+3W(gp)\,[2U^{BC}(gp)]^2(m^C)^2\\
&\times\;(6m^A m^C+2m^B-1)-3W(gp)^2 2U^{BC}(gp)(m^C)^2\,[6m^A(1-m^A)-1])\Big]-[(m^A m^C)^2/(1-m^A)]\,(\Gamma^{AA}(gp)m^A\\
&\times\{W(gp)2U^{AB}(gp)[6m^A(1-m^A)-1]+2U^{AB}(gp)\,2U^{BC}(gp)\,m^C(1-4m^A)+W(gp)^2m^C(1-4m^A)+m^C(1-2m^C)\;W(gp)2U^{BC}\\
&\times(gp)\}+m^A\Gamma^{AC}(gp)\,\{m^C(1-2m^A)\,2U^{AB}(gp)2U^{BC}(gp)+m^C(1-2m^A)W(gp)^2+m^C(1-4m^C)W(gp)2V^{BC}(gp)\}\\
&-\tfrac{1}{2}(m^C-m^B)\,\{W(gp)^2 2U^{AB}(gp)(1-m^A)\,[6m^A(1-m^A)-1]+2U^{AB}(gp)\,[2U^{BC}(gp)]^2m^C(6m^A m^C+2m^B-1)\\
&-\;W(gp)2U^{AB}(gp)2U^{BC}(gp)\;2m^C[6m^A(1-m^A)-1]-W(gp)^3m^C[6m^A(1-m^A)-1]\\
&-W(gp)\,[2U^{BC}(gp)]^2m^C[6m^C(1-m^C)-1]+W(gp)^2 2U^{BC}(gp)\;2m^C(6m^A m^C+2m^B-1)\})\\
&+[m^A m^C/(1-n^C)]^2\Big[m^A m^C\,\Gamma^{AC}(gp)\,\{[6m^A(1-m^A)-1]\,[2U^{AB}(gp)]^2+2m^C(1-4m^A)W(gp)2U^{AB}(gp)\\
&+m^C(1-2m^C)W(gp)^2\}+m^A m^C\,\Gamma^{CC}(gp)\,[2m^C(1-2m^A)W(gp)\;2U^{AB}(gp)+m^C(1-4m^C)W(gp)^2]+\tfrac{1}{2}(m^B-m^A)\\
&\times(\Gamma^{AC}(gp)\,\{[6m^A(1-m^A)-1]\,[2U^{AB}(gp)]^2+m^C(1-2m^C)W(gp)^2\;+2m^C(1-4m^A)W(gp)2U^{AB}(gp)\}\\
&+\Gamma^{CC}(gp)[m^C(1-4m^C)\;W(gp)^2+2m^C(1-2m^A)W(gp)2U^{AB}(gp)|\;+[2U^{AB}(gp)]^3m^A(1-m^A)[6m^A(1-m^A)-1]\\
&-W(gp)^3\;\;m^A m^C[6m^C(1-m^C)-1]+3W(gp)^2\,2U^{AB}(gp)m^A m^C\;\;(6m^A m^C+2m^B-1)\\
&-3W(gp)\,[2U^{AB}(gp)]^2\,m^A m^C[6m^A(1-m^A)-1])\Big]+(m^A)^2m^C(m^C\,\Gamma^{AC}(gp)\,\{W(gp)^2m^C(1-4m^A)\\
&+W(gp)2U^{AB}(gp)[6m^A(1-m^A)-1]+W(gp)2U^{BC}(gp)m^C(1-2m^C)\;\;+2U^{AB}(gp)\,2U^{BC}(gp)m^C(1-4m^A)\}\\
&+m^C\,\Gamma^{CC}(gp)\,\{W(gp)^2m^C(1-2m^A)+m^C(1-2m^A)\,2U^{AB}(gp)2U^{BC}(gp)+m^C(1-4m^C)W(gp)2U^{BC}(gp)\}\\
&+\tfrac{1}{2}(m^B-m^A)\,\{W(gp)^3m^C(6m^A m^C+2m^B-1)+W(gp)\;[2U^{AB}(gp)]^2(1-m^A)\,[6m^A(1-m^A)-1]\\
&-2m^C\,[6m^A(1-m^A)-1]\;\;W(gp)^2 2U^{AB}(gp)-2U^{BC}(gp)\,[2U^{AB}(gp)]^2m^C[6m^A(1-m^A)-1]\\
&-W(gp)^2\,2U^{BC}(gp)\,m^C[6m^C(1-m^C)-1]+2m^C(6m^A m^C+2m^B-1)\,W(gp)2U^{AB}(gp)\;2U^{BC}(gp)\})\Big\}\quad,
\end{aligned}
\tag{A4}
$$

$$\Gamma^{AC}(gp) \equiv U^{AC}(gp)(2m^A m^C + m^B) + U^{AB}(gp)m^B(2m^A - 1) + U^{BC}(gp)m^B(2m^C - 1) , \tag{A5}$$

$$\Gamma^{AA}(gp) \equiv U^{AB}(gp)2m^B(m^A - 1) + U^{AC}(gp)2m^C(m^A - 1) + U^{BC}(gp)\, 2m^B m^C , \tag{A6}$$

$$\Gamma^{CC}(gp) \equiv U^{BC}(gp)2m^B(m^C - 1) + U^{AC}(gp)\, 2m^A(m^C - 1) + U^{AB}(gp)2m^A m^B . \tag{A7}$$

APPENDIX B

The values of $\Psi_i(\vec{k})$ can be expressed in terms of the Fourier transforms of the $\Phi_i(\overrightarrow{gp})$, i.e.,

$$\Phi_i(\vec{g}\vec{p}) = \frac{1}{N}\sum_{\vec{k}} \Phi_i(\vec{k})e^{i\vec{k}\cdot(\vec{g}-\vec{p})} , \tag{B1}$$

$$\Phi_i(\vec{k}) = \sum_{\vec{g}\vec{p}} \Phi_i(\vec{g}\vec{p})e^{-i\vec{k}\cdot(\vec{g}-\vec{p})} . \tag{B2}$$

These relationships are

$$\Psi_1^{AC}(\vec{k}) = (1/m^A m^C)\,\Phi_1^{AC}(\vec{k}) , \tag{B3}$$

$$\Psi_2^{AC}(\vec{k}) = (1/m^A m^C)^2\,[m^A m^C \Phi_2^{AC}(\vec{k}) + \Phi_1^{AC}(\vec{k})^2] , \tag{B4}$$

$$\Psi_3^{AC}(\vec{k}) = (1/m^A m^C)^3\,[(m^A m^C)^2 \Phi_3^{AC}(\vec{k}) + 2m^A m^C \Phi_1^{AC}(\vec{k})\,\Phi_2^{AC}(\vec{k}) + \Phi_1^{AC}(\vec{k})^3] . \tag{B5}$$

APPENDIX C

The appropriate spin subspace in which to treat the three-level system would be that of spin one $(2S+1=3)$. Using the null operator,

$$(S_i^z - 1)\,S_i^z\,(S_i^z + 1) = 0 , \tag{C1}$$

we can assign occupation operators to each spin state, i.e.,

$$\sigma_i^A \rightarrow aS_i^z(S_i^z + 1) , \tag{C2a}$$

$$\sigma_i^B \rightarrow b(S_i^z - 1)(S_i^z + 1) , \tag{C2b}$$

$$\sigma_i^C \rightarrow cS_i^z(S_i^z - 1) . \tag{C2c}$$

The normalization constants a, b, and c are determined from the properties of the σ_i^λ, i.e.,

$$\sigma_i^\lambda \sigma_j^\lambda = 0 \quad (i \neq j) , \tag{C3a}$$

$$\sigma_i^\lambda \sigma_i^\nu = 0 \quad (\lambda \neq \nu) , \tag{C3b}$$

$$\sigma_i^\lambda \sigma_i^\lambda = \sigma_i^\lambda , \tag{C3c}$$

from which Eq. (2.2) follows.

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[1] R. A. Tahir-Kheli, Phys. Rev. 169, 517 (1968).

[2] R. A. Tahir-Kheli, Phys. Rev. 188, 1142 (1969); D. H. Wu and R. A. Tahir-Kheli (unpublished).

[3] Note that in order to obtain the two-point correlation function $\langle \sigma_g^A \sigma_p^C \rangle$ to the given accuracy [see Eq. (2.14)], we need to know five-point correlations of the type $\langle \sigma_{f_1}^{\lambda_1} \ldots \sigma_{f_5}^{\lambda_5} \rangle$ correct to the order $(1/T)^0$, four-point correlations $\langle \sigma_{f_1}^{\lambda_1} \ldots \sigma_{f_4}^{\lambda_4} \rangle$ to the order $(1/T)$, three-point correlations $\langle \sigma_{f_1}^{\lambda_1} \sigma_{f_2}^{\lambda_2} \sigma_{f_3}^{\lambda_3} \rangle$ to the order $(1/T)^2$, and two-point correlations of the form $\langle \sigma_{f_1}^{\lambda_1} \sigma_{f_2}^{\lambda_2} \rangle$ to the order $(1/T)^3$, and of course, the functions $X(0)$ and $Y(0)$ correct to the order $(1/T)^3$. (Note there that while the various locations, f_i are all to be different, the corresponding lambda's λ_i are arbitrary.) The expressions for these correlations are very lengthy and are given in G. B. Taggart, Ph.D. dissertation, Temple University, 1970 (unpublished). The only reason for writing out these correlation functions somewhere in the literature is that if the SRO parameters were to be calculated to the next order, i.e., $O(1/T)^4$, these expressions would again be needed with one additional order of accuracy.

[4] For a discussion of the procedure to be followed for the calculation of the transition temperature from the knowledge of $\alpha^{\mu\nu}(\vec{k})$, refer to I and to Ref. 2. The important point to note here is that in contradiction to the impression created by I, this procedure leads to only *one* result for the transition temperature. For the present purposes this transition temperature is defined to be that temperature at which the system would be expected to first show any long-range order as the system temperature is reduced from $T=\infty$. In this connection it should be noted that the predictions of I and of this work regarding the type of long-range order that first sets in are to be understood as follows: For given composition and interparticle potentials, if say $(k_BT_c)^{\lambda'\nu'}$, is the largest of the three results $(k_BT_c)^{\lambda\nu}$, where λ, $\nu = A, B, C$, then the binary long-range order that first sets in would be of the variety $\lambda'\nu'$. Note also that this interpretation does not exclude the situation in which the ternary ordering itself separates the disordered and the ordered phases. For this case all the three expressions for $(k_BT_c)^{\lambda\nu}$ would, of course, be the same. Note also that the present work makes no predictions regarding the structure of the ordering below the temperature $(k_BT_c)^{\lambda'\nu'}$ at which some binary ordering $\lambda'\nu'$ first appears. It could be that at lower temperatures some other binary ordering, or even some type of ternary ordering, would be more stable. Moreover, the stability of these orderings would also depend upon the existence of higher-body potentials and also, of course, upon the dynamics, which has been completely ignored in the present discussion.