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An Extension of the Coherent-Potential Approximation. II

——*Single-Particle Excitation Functions*——

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On the basis of an extension of the coherent-potential approximation, the single-particle excitation function of a quasi-particle in a substitutionally disordered system is formulated for the Hamiltonian with off-diagonal disorder in addition to diagonal one. A particular attention is paid to the effect of site-dependence of the strength of coupling between a quasi-particle and an external quantum. The formalism is applicable to the case where the inverse of the propagator of the quasi-particle is a quasi-additive random operator. The theory is compared with other existing theories which have their own ranges of applicability.

§ 1. Introduction

The coherent-potential approximation (CPA) invented by Soven[1] and Taylor[2] is a great progress in the theoretical investigation of motion of quasi-particles in substitutionally disordered systems. The essence of CPA is that the average medium for a quasi-particle is determined self-consistently within the single-site approximation. The applicability of CPA in its original form is limited to the case where the inverse of the propagator of the quasi-particle has a diagonal disorder but no off-diagonal disorder of any kind. Therefore various attempts[3)~6)] to extend the range of applicability were proposed. We have shown in a previous paper[3] (referred to as I hereafter) that the renormalization-propagator formalism (RPF) proposed there and the formalism due to Blackmann et al.[6] (referred to as BEB hereafter) are two extremes of extensions within the self-consistent single-site approximation (SCSSA). We have concentrated in I exclusively upon the total and partial densities of states. It is also desirable to establish the formalism for the calculation of other observables of interest, e.g., the single-particle excitation functions (SPEF) and the transport coefficients. The aim of the present paper is to present a formalism of evaluating SPEF within SCSSA.

The SPEF gives us the rate of such an elementary process that a quasi-particle is created or annihilated at the moment when a quantum such as a photon or a neutron impinging into the material is absorbed or scattered. It is a function of the energy and the momentum transferred to the material from the quantum. SPEF in a disordered material is very important quantity since it has direct information about the state of quasi-particles in the material. There are various

examples[7] of SPEF of disordered materials, e.g., the one-phonon neutron scattering intensity of a disordered lattice, the one-magnon neutron scattering intensity of a mixed Heisenberg ferromagnet, the optical absorption coefficient of a mixed dielectric crystal in which Frenkel excitons may be excited, the Raman scattering intensity of a mixed dielectric crystal having optical branches of phonon, the infrared absorption coefficient of a mixed polar crystal, etc.

Let us describe here the general structure of SPEF in a substitutionally disordered system.[7] In such a system, the coupling constant between a quasi-particle and a quantum of external origin fluctuates from site to site since different nucleus, atom or ion may have different neutron scattering length, different transition dipole moment, different effective charge, etc. Therefore SPEF in a disordered system has a bilinear form with respect to coupling constants associated with the component nuclei, atoms, ions, etc. The coefficients of the bilinear form are proportional to the partial spectral densities, i.e., the imaginary part of the Fourier transform of the configuration averaged partial propagators each of which is associated with propagation of the quasi-particle between two sites when the type of the nucleus, atom, or ion occupying each site is specified.

It is usually impossible to evaluate exactly the configuration average of the partial propagators as well as that of the propagator itself. Now we have CPA and its various extensions[3)~6)] which allow us to evaluate the averaged propagator within SCSSA. Therefore it is desirable to have a method of evaluating the averaged partial propagators within the same approximation. Elliott and Taylor[8] showed that the averaged partial propagators can be related to the averaged propagator if the system is a binary disordered system and the inverse of the propagator is a diagonal random operator.*) Their method cannot be generalized, however, to the case where the propagator does not satisfy these conditions. On the other hand, what are obtained directly in BEB are the averaged partial propagators themselves and the averaged propagator is rather a secondary quantity being derived from those. Therefore SPEF in a disordered system can be evaluated by using BEB within its range of applicability, i.e., the case where the inverse of the propagator is a canonical random operator.*) We propose in the present paper another method which allows us to evaluate the averaged partial propagators within SCSSA when the inverse of the propagator is an additive random operator or a quasi-additive one. It can be shown that BEB and the present method are equivalent not only to each other within the common range of applicability but also to the method of Elliott and Taylor for the propagator whose inverse is a diagonal random operator.

The present paper is organized as follows: In § 2 we review the formalism, developed in I, of evaluating in SCSSA the configurational average of the propagator whose inverse is an additive random operator. In § 3 we present in the

*) Exact prescriptions of various types of random operators appearing here and to appear in what follows are presented in I.

first place an exact definition of the partial spectral densities. We next investigate how they change when the propagator is renormalized. Then, we undertake to evaluate in SCSSA configurational averages of the partial propagators. In § 4 we discuss a number of subjects associated with our formalism.

§ 2. Evaluation of the averaged propagator

We take for definiteness a binary mixed crystal A_xB_y in which two species of atoms A and B occupy randomly with probabilities x and y $(x+y=1)$ the lattice sites of a Bravais lattice. Also we assume that the quasi-particle concerned has only one degree of freedom per site; the result can be easily generalized to the case where the quasi-particle has several degrees of freedom per site, e.g., the case of phonon.

The propagator of the quasi-particle takes usually the following form:[3]

$$\mathcal{G}(z)=(\eta_{\mathrm{op}}z-\mathcal{K})^{-1}, \tag{2·1}$$

where z is a complex variable representing "energy" of the quasi-particle, $\mathcal{K}$ a random operator including the site-energy and the transfer-energy of the quasi-particle and η_{op} the "metric operator". In the site-representation, $\mathcal{K}$ is real symmetric matrix and η_{op} takes the form

$$\eta_{\mathrm{op}}=\sum_n |n\rangle\eta_n\langle n|, \tag{2·2}$$

where η_n is a random variable taking a positive number η^A or η^B. The propagator has an important symmetry with respect to the complex conjugate operation in the site-representation, namely, $\overline{\mathcal{G}(z)}=\mathcal{G}(\bar{z})$, which assures the time reversal invariance of the motion of the quasi-particle.

The averaged propagator and the conditionally averaged propagators are defined by

$$G(z)\equiv\langle\mathcal{G}(z)\rangle, \tag{2·3}$$

$$\mathcal{G}^i(n:z)\equiv\langle\mathcal{G}(z)\rangle^{i/n}, \quad (i=A, B) \tag{2·4}$$

where $\langle\cdots\rangle$ denotes the configurational average and $\langle\cdots\rangle^{i/n}$ the conditional configurational average under the condition that site n is occupied by an atom of type i. The conditionally averaged propagators can be regarded as two possible values of a random operator $\mathcal{G}(n:z)$, which is also referred to by the same name hereafter. It follows that

$$G(z)=\langle\mathcal{G}(n:z)\rangle\equiv\sum_i c^i\mathcal{G}^i(n:z), \tag{2·5}$$

where $c^A\equiv x$ and $c^B\equiv y$.

We assume that $\mathcal{K}$ in Eq. (2·1) is an additive random operator. Then $\mathcal{G}(z)$ is written as

$$\mathcal{G}(z)=(\mathcal{D}_{\mathrm{op}}(z)-H_0)^{-1}, \tag{2·6}$$

$$\mathcal{D}_{\text{op}}(z)=\sum_n \mathcal{D}(n:z), \tag{2·7}$$

where H_0 is a periodic operator being real and symmetric in the site-representation and $\mathcal{D}(n:z)$ is a random operator taking $\mathcal{D}^A(n:z)$ or $\mathcal{D}^B(n:z)$. We denote by $\mathfrak{S}_n$ the smallest sub-Hilbert space of the single-quasi-particle Hilbert space among those to which both of $\mathcal{D}^A(n:z)$ and $\mathcal{D}^B(n:z)$ can be confined. Let ν be the dimension of $\mathfrak{S}_n$ and let $\{|n,1\rangle, |n,2\rangle, \cdots, |n,\nu\rangle\}$ be a complete set of real basis vectors in $\mathfrak{S}_n$.*) Then we have

$$\mathcal{D}^i(n:z)=\|n\rangle\mathcal{D}^i(z)\langle n\|, \quad (i=A,B) \tag{2·8}$$

where $\mathcal{D}^i(z)$ $(i=A,B)$ are ν-dimensional symmetric matrices, $\|n\rangle\equiv(|n,1\rangle, |n,2\rangle, \cdots, |n,\nu\rangle)$ is a "ket" and $\langle n\|$ is the corresponding "bra", i.e., a ν-dimensional column vector whose elements are the ordinary bras: $\langle n,1|, \langle n,2|, \cdots, \langle n,\nu|$.

According to *I*, $G(z)$ and $\mathcal{G}^i(n:z)$ $(i=A,B)$ are given under SCSSA by

$$G(z)=(\sum_n \|n\rangle \boldsymbol{D}(z)\langle n\| - H_0)^{-1}, \tag{2·9}$$

$$\mathcal{G}^i(n:z)=\{G(z)^{-1}+\|n\rangle(\mathcal{D}^i(z)-\boldsymbol{D}(z))\langle n\|\}^{-1}, \quad (i=A,B) \tag{2·10}$$

where $\boldsymbol{D}(z)$ is a ν-dimensional symmetric matrix determined by the condition that $G(z)$ and $\mathcal{G}^i(n:z)$ given by Eqs. (2·9) and (2·10) satisfy the self-consistency condition equation (2·5).

The ν-dimensional symmetric matrices defined by the equations

$$\boldsymbol{F}(z)\equiv\langle n\|G(z)\|n\rangle, \tag{2·11}$$

$$\boldsymbol{F}^i(z)\equiv\langle n\|\mathcal{G}^i(n:z)\|n\rangle \quad (i=A,B) \tag{2·12}$$

satisfy the equations

$$\boldsymbol{F}(z)=\sum_i c^i\boldsymbol{F}^i(z), \quad \text{(cf. Eq. (2·5))} \tag{2·13}$$

$$\boldsymbol{F}^i(z)=\{\boldsymbol{F}(z)^{-1}+\boldsymbol{D}^i(z)-\boldsymbol{D}(z)\}^{-1}. \quad (i=A,B) \tag{2·14}$$

Using a Bloch state $|\boldsymbol{k}\rangle\equiv\sum_n \exp(i\boldsymbol{k}\cdot\boldsymbol{R}_n)|n\rangle$ ($\boldsymbol{R}_n$ being the lattice vector for site n), we define a ν-dimensional row vector $\langle\boldsymbol{k}\|0\rangle$ by the equation

$$\langle\boldsymbol{k}\|0\rangle=(\langle\boldsymbol{k}|0,1\rangle, \langle\boldsymbol{k}|0,2\rangle, \cdots, \langle\boldsymbol{k},|0,\nu\rangle), \tag{2·15}$$

where 0 denotes a reference site chosen arbitrarily. Then the Fourier transform of $G(z)$ is given by

$$G(\boldsymbol{k}:z)=\{\langle\boldsymbol{k}\|0\rangle\boldsymbol{D}(z)\langle 0\|\boldsymbol{k}\rangle-\varepsilon_0(\boldsymbol{k})\}^{-1}, \tag{2·16}$$

where the column vector $\langle 0\|\boldsymbol{k}\rangle$ is the hermitean conjugate to $\langle\boldsymbol{k}\|0\rangle$ and $\varepsilon_0(\boldsymbol{k})$ is the Fourier transform of H_0. $\boldsymbol{F}(z)$ is written in terms of $G(\boldsymbol{k}:z)$ as

$$\boldsymbol{F}(z)=\int_{\boldsymbol{k}}\langle 0\|\boldsymbol{k}\rangle G(\boldsymbol{k}:z)\langle\boldsymbol{k}\|0\rangle, \tag{2·17}$$

*) Here a real vector is meant to be a state vector represented by a real column vector in the site-representation. Time reversal invariance allows us to choose the basis vectors of $\mathfrak{S}_n$ as real vectors.

where $\int_{\boldsymbol{k}}$ denotes an operation of averaging a $\boldsymbol{k}$-dependent function over the first Brillouin zone in the $\boldsymbol{k}$-space. Equations (2·16) and (2·17) show that each matrix element of $\boldsymbol{F}(z)$ is a function of $\boldsymbol{D}(z)$. Therefore Eq. (2·13) together with Eqs. (2·14) provides a self-consistency condition on $\boldsymbol{D}(z)$.

§ 3. Evaluation of the averaged partial propagators

In order to give a precise definition of the partial propagators, let us introduce occupation indices[7] $\chi_n{}^i$ $(i=A, B)$ which are random variables taking 1 or 0 according to whether the type of the atom occupying site n is the one specified by the index i or not, respectively. The partial propagators $\mathcal{G}^{ij}(z)$ $(i, j=A, B)$ are then defined by the equations

$$\mathcal{G}^{ij}(z) \equiv \chi_{\mathrm{op}}^{i} \mathcal{G}(z) \chi_{\mathrm{op}}^{j}, \quad (i, j=A, B) \tag{3·1}$$

where

$$\chi_{\mathrm{op}}^{i} \equiv \sum_{n} |n\rangle \chi_n{}^i \langle n| \quad (i=A, B) \tag{3·2}$$

are projection operators each of which projects the single-quasi-particle Hilbert space on to a subspace spanned by all the basis vectors localized on the sites occupied by atoms of the type denoted by its index. Noticing the obvious identity

$$\sum_{i} \chi_{\mathrm{op}}^{i} = 1_{\mathrm{op}}, \tag{3·3}$$

we see the following identity:

$$\mathcal{G}(z) = \sum_{i, j} \mathcal{G}^{ij}(z). \tag{3·4}$$

Therefore the averaged partial propagators

$$G^{ij}(z) \equiv \langle \mathcal{G}^{ij}(z) \rangle \quad (i, j=A, B) \tag{3·5}$$

satisfy

$$G(z) = \sum_{i, j} G^{ij}(z). \tag{3·6}$$

Similarly the spectral density and the partial spectral densities defined by

$$\sigma(\boldsymbol{k}:E) \equiv -\frac{1}{\pi} \operatorname{Im} G(\boldsymbol{k}:E+i0), \tag{3·7}$$

$$\sigma^{ij}(\boldsymbol{k}:E) \equiv -\frac{1}{\pi} \operatorname{Im} G^{ij}(\boldsymbol{k}:E+i0), \quad (i, j=A, B) \tag{3·8}$$

respectively, ($G^{ij}(\boldsymbol{k}:z)$ being the Fourier transform of $G^{ij}(z)$ $(i, j=A, B)$) satisfy

$$\sigma(\boldsymbol{k}:E) = \sum_{i, j} \sigma^{ij}(\boldsymbol{k}:E). \tag{3·9}$$

SPEF for the quasi-particle is proportional to

$$\Phi(\boldsymbol{k}:E) = \sum_{i, j} \gamma^i \gamma^j \sigma^{ij}(\boldsymbol{k}:E), \tag{3·10}$$

where γ^i $(i=A, B)$ are the two coupling constants of the interaction between the quasi-particle and the quantum of external origin. Let $\hbar\boldsymbol{q}$ be the momentum transferred from the quantum to the system. Then the wave vector $\boldsymbol{k}$ of SPEF is equal to the wave vector derived by reducing $\boldsymbol{q}$ to the first Brillouin zone.

From the time reversal invariance we obtain $G^{ij}(\boldsymbol{k}:z)=G^{ji}(-\boldsymbol{k}:z)$ $(i,j=A, B)$ and hence $\sigma^{ij}(\boldsymbol{k}:E)=\sigma^{ji}(-\boldsymbol{k}:E)$ $(i,j=A, B)$. Therefore $G(\boldsymbol{k}:z)$, $\sigma(\boldsymbol{k}:E)$ and $\boldsymbol{\Phi}(\boldsymbol{k}:E)$ are all even functions of $\boldsymbol{k}$. If the system has the inversion symmetry (of course after averaged), $G^{ij}(\boldsymbol{k}:E)$ and $\sigma^{ij}(\boldsymbol{k}:E)$ $(i,j=A, B)$ are even functions of $\boldsymbol{k}$ and hence symmetric with respect to interchange of the indices i and j. Since we have assumed a Bravais lattice, we may assume that the system has the inversion symmetry.

If the inverse of the propagator $\mathcal{G}(z)$ is a quasi-additive random operator, we may renormalize[3] the propagator as $\hat{\mathcal{G}}(z)\equiv\xi_{\mathrm{op}}\mathcal{G}(z)\xi_{\mathrm{op}}$ in such a way that the inverse of the renormalized propagator $\hat{\mathcal{G}}(z)$ is an additive random operator, where ξ_{op} is a diagonal random operator defined in terms of the two non-zero real numbers ξ^A and ξ^B by $\xi_{\mathrm{op}}=\xi^A\chi^A_{\mathrm{op}}+\xi^B\chi^B_{\mathrm{op}}$. We may define the renormalized partial spectral densities $\hat{\sigma}^{ij}(\boldsymbol{k}:E)$ $(i,j=A,B)$ in terms of the renormalized propagator in a similar way as we have defined the partial spectral densities in terms of the propagator before renormalized. It is an easy task to show that they satisfy $\hat{\sigma}^{ij}(\boldsymbol{k}:E)=\xi^i\xi^j\sigma^{ij}(\boldsymbol{k}:E)$ $(i,j=A, B)$. Therefore $\boldsymbol{\Phi}(\boldsymbol{k}:E)$ given by Eq. (3·10) is given also by a similar equation but in terms of the renormalized spectral densities and the renormalized coupling constants $\hat{\gamma}^i\equiv\gamma^i/\xi^i$ $(i=A, B)$. Thus SPEF for a quasi-particle with a propagator whose inverse is a quasi-additive random operator is shown to be reduced by the renormalization trick to SPEF for that but with a propagator whose inverse is an additive random operator. Hence we assume in what follows that the inverse of the propagator is an additive random operator.

We now enter on the main presentation of the paper. The wave matrix defined by the equation

$$\Omega_{\mathrm{op}}(z)\equiv G(z)^{-1}\mathcal{G}(z) \tag{3·11}$$

satisfies

$$\langle\Omega_{\mathrm{op}}(z)\rangle=1_{\mathrm{op}}\,. \tag{3·12}$$

The wave matrix has a function of extracting from a random operator a part being coherent with the random operator $\mathcal{G}(z)$. The coherent parts of the random operators χ^i_{op} $(i=A, B)$ are given by

$$X^i_{\mathrm{op}}(z)=\langle\Omega_{\mathrm{op}}(z)\chi^i_{\mathrm{op}}\rangle\,. \quad (i=A, B) \tag{3·13}$$

It follows that

$$\sum_i X^i_{\mathrm{op}}(z)=1_{\mathrm{op}}. \quad \text{(cf., Eq. (3·3))} \tag{3·14}$$

The corresponding incoherent parts

$$\chi^i_{\rm inc}(z) \equiv \chi^i_{\rm op} - X^i_{\rm op}(z) \quad (i=A,B) \tag{3·15}$$

satisfy

$$\langle \mathcal{G}(z)\chi^i_{\rm inc}(z)\rangle = 0\,. \quad (i=A,B) \tag{3·16}$$

It should be noted that $X^i_{\rm op}(z)$ and $\chi^i_{\rm inc}(z)$ $(i=A,B)$ are not site-diagonal in contrast to $\chi^i_{\rm op}$ $(i=A,B)$ and besides these, as well as $\Omega_{\rm op}(z)$, are not symmetric in the site-representation.

Transposing Eqs. (3·16) yields

$$\langle \chi^i_{\rm inc}(z)^T \mathcal{G}(z)\rangle = 0\,, \quad (i=A,B) \tag{3·17}$$

where the superscript T denotes the transpose operation in the site-representation. From Eqs. (3·16) and (3·17) we obtain

$$\begin{aligned} G^{ij}(z) &= \langle (X^i_{\rm op}(z)^T + \chi^i_{\rm inc}(z)^T)\,\mathcal{G}(z)\,(X^j_{\rm op}(z) + \chi^j_{\rm inc}(z))\rangle \\ &= X^i_{\rm op}(z)^T G(z)\, X^j_{\rm op}(z) + G^{ij}_{\rm inc}(z)\,, \quad (i,j=A,B) \end{aligned} \tag{3·18}$$

where

$$G^{ij}_{\rm inc}(z) \equiv \langle \chi^i_{\rm inc}(z)^T \mathcal{G}(z)\chi^j_{\rm inc}(z)\rangle\,. \quad (i,j=A,B) \tag{3·19}$$

Inserting Eqs. (3·2) into Eqs. (3·13) yields

$$X^i_{\rm op}(z) = \sum_n X^i(n:z)\,, \quad (i=A,B)\,, \tag{3·20}$$

$$X^i(n:z) = c^i \Omega^i(n:z)|n\rangle\langle n|\,, \quad (i=A,B) \tag{3·21}$$

where

$$\Omega^i(n:z) \equiv \langle \Omega_{\rm op}(z)\rangle^{i/n} = G(z)^{-1}\mathcal{G}^i(n:z)\,. \quad (i=A,B) \tag{3·22}$$

Hence $X^i_{\rm op}(z)$ $(i=A,B)$ are given in terms of the quantities which can be evaluated within SCSSA as shown in § 2. Then, in order to obtain the averaged partial propagators $G^{ij}(z)$ $(i,j=A,B)$ in SCSSA, we have to evaluate $G^{ij}_{\rm inc}(z)$ $(i,j=A,B)$ in the same approximation.

Now $\Omega^i(n:z)$ $(i=A,B)$ can be regarded as two possible values of the random operator:

$$\Omega(n:z) \equiv G(z)^{-1}\mathcal{G}(n:z)\,, \quad \text{(cf., Eq. (3·11))} \tag{3·23}$$

which satisfies

$$\langle \Omega(n:z)\rangle = \sum_i c^i \Omega^i(n:z) = 1_{\rm op}\,. \quad \text{(cf., Eq. (3·12))} \tag{3·24}$$

The quantities $X^i(n:z)$ $(i=A,B)$ can be regarded as the coherent parts of the random operators:

$$\chi^i(n) \equiv |n\rangle \chi_n{}^i \langle n|\,. \quad (i=A,B) \tag{3·25}$$

Indeed Eqs. (3·21) can be written as

$$X^i(n:z) = \langle \Omega(n:z)\chi^i(n)\rangle\,. \quad (i=A,B) \tag{3·26}$$

The corresponding incoherent parts

$$\chi^i_{\rm inc}(n:z)\equiv\chi^i(n)-X^i(n:z) \quad (i=A,B) \tag{3·27}$$

satisfy the equations

$$\left.\begin{aligned}&\langle\mathcal{G}(z)\chi^i_{\rm inc}(n:z)\rangle=\langle\mathcal{G}(n:z)\chi^i_{\rm inc}(n:z)\rangle=0\,, \quad (i=A,B)\\ &\langle\chi^i_{\rm inc}(n:z)^T\mathcal{G}(z)\rangle=\langle\chi^i_{\rm inc}(n:z)^T\mathcal{G}(n:z)\rangle=0\,. \quad (i=A,B)\end{aligned}\right\} \tag{3·28}$$

Inserting the equations

$$\chi^i_{\rm inc}(z)=\sum_n\chi^i_{\rm inc}(n:z) \quad (i=A,B) \tag{3·29}$$

and their transposed equations into Eqs. (3·19) yields

$$G^{ij}_{\rm inc}(z)=\sum_{n,n'}\langle\chi^i_{\rm inc}(n:z)^T\mathcal{G}(z)\chi^j_{\rm inc}(n':z)\rangle. \quad (i,j=A,B) \tag{3·30}$$

Noting Eqs. (3·28) we may neglect under SCSSA the cross terms in the summand of the double summations in Eqs. (3·30) and obtain

$$G^{ij}_{\rm inc}(z)=\sum_n G^{ij}_{\rm inc}(n;z), \quad (i,j=A,B) \tag{3·31}$$

$$G^{ij}_{\rm inc}(n:z)=\langle\chi^i_{\rm inc}(n:z)^T\mathcal{G}(n:z)\chi^j_{\rm inc}(n:z)\rangle\,. \quad (i,j=A,B) \tag{3·32}$$

By making use of Eqs. (3·23), (3·26) and (3·27), Eqs. (3·32) can be written as

$$\begin{aligned}&G^{ij}_{\rm inc}(n:z)\\ &=\langle\chi^i(n)\mathcal{G}(n:z)\chi^j(n)\rangle-X^i(n:z)^TG(z)X^j(n:z) \quad (i,j=A,B)\ \text{(cf., Eq.(3·18))}\\ &=\delta_{ij}c^i|n\rangle F^i(z)\langle n|-X^i(n:z)^TG(z)X^j(n:z), \quad (i,j=A,B)\end{aligned} \tag{3·33}$$

where

$$F^i(z)\equiv\langle n|\mathcal{G}^i(n:z)|n\rangle. \quad (i=A,B) \tag{3·34}$$

Equations (3·31), (3·33) and (3·34) together with Eqs. (3·21) and (3·22) show that $G^{ij}_{\rm inc}(z)$ $(i,j=A,B)$ can also be given in terms of the quantities which can be evaluated within SCSSA. Thus we have solved the problem of evaluating the averaged partial propagators within SCSSA.

We will evaluate the Fourier transforms of the averaged partial propagators. Equations (2·10) and (3·22) yield

$$\mathcal{Q}^i(n:z)=1_{\rm op}-\|n\rangle(\mathcal{D}^i(z)-\boldsymbol{D}(z))\langle n\|\mathcal{G}^i(n:z). \quad (i=A,B) \tag{3·35}$$

Whence we have

$$\mathcal{Q}^i(n:z)\|n\rangle=\|n\rangle\boldsymbol{\mathcal{Q}}^i(z), \quad (i=A,B) \tag{3·36}$$

where $\boldsymbol{\mathcal{Q}}^i(z)$ $(i=A,B)$ are ν-dimensional matrices with various expressions ($\mathbf{1}$ being the ν-dimensional unit matrix):

$$\boldsymbol{\mathcal{Q}}^i(z)=\mathbf{1}-(\mathcal{D}^i(z)-\boldsymbol{D}(z))\boldsymbol{F}^i(z), \quad (i=A,B) \tag{3·37}$$

$$=\{\mathbf{1}+(\mathcal{D}^i(z)-\boldsymbol{D}(z))\boldsymbol{F}(z)\}^{-1}, \quad (i=A,B) \tag{3·38}$$

$$=\boldsymbol{F}(z)^{-1}\boldsymbol{F}^{i}(z). \quad (i=A, B) \tag{3·39}$$

As a result of the self-consistency condition equation (2·13), we have

$$\sum_{i} c^{i}\boldsymbol{\Omega}^{i}(z)=\mathbf{1}. \quad \text{(cf., Eq. (3·24))} \tag{3·40}$$

Now we may choose the set of basis vectors of $\mathfrak{S}_n$ in such a way that $|n, 1\rangle=|n\rangle$. Then the first diagonal elements of $\boldsymbol{F}^{i}(z)$ $(i=A, B)$ are equal to $F^{i}(z)$ $(i=A, B)$ (see Eqs. (3·34)). Then from Eqs. (3·21), (3·36) and (3·39) we can write Eqs. (3·33) as

$$G^{ij}_{\mathrm{inc}}(n:z)=|n\rangle S^{ij}(z)\langle n|, \quad (i, j=A, B) \tag{3·41}$$

$$S^{ij}(z)=\{\delta_{ij}c^{i}\boldsymbol{F}^{i}(z)-c^{i}\boldsymbol{F}^{i}(z)\boldsymbol{F}(z)^{-1}c^{j}\boldsymbol{F}^{j}(z)\}_{1,1}, \quad (i, j=A, B) \tag{3·42}$$

where $\{\cdots\}_{1,1}$ denotes the first diagonal element of the matrix in the brackets. It should be noticed that $G^{ij}_{\mathrm{inc}}(n:z)$ $(i, j=A, B)$ are localized on site n. It follows that

$$G^{ij}_{\mathrm{inc}}(z)=S^{ij}(z)1_{\mathrm{op}}. \quad (i, j=A, B) \tag{3·43}$$

Therefore we have

$$G^{ij}(\boldsymbol{k}:z)=X^{i}(\boldsymbol{k}:z)G(\boldsymbol{k}:z)X^{j}(\boldsymbol{k}:z)+S^{ij}(z), \quad (i, j=A, B) \tag{3·44}$$

where $X^{i}(\boldsymbol{k}:z)$ $(i=A, B)$ are the Fourier transforms of $X^{i}_{\mathrm{op}}(z)$ $(i=A, B)$ and also of $X^{i}_{\mathrm{op}}(z)^{T}$ $(i=A, B)$ owing to the time reversal invariance and the inversion symmetry as shown later on. Equations (3·20), (3·21) and (3·36) yield

$$\begin{aligned} X^{i}(\boldsymbol{k}:z)&=\langle\boldsymbol{k}|X^{i}(0:z)|\boldsymbol{k}\rangle=c^{i}\langle\boldsymbol{k}|\Omega^{i}(0:z)|0\rangle \\ &=\sum_{\alpha=1}^{\nu}\langle\boldsymbol{k}|0, \alpha\rangle c^{i}\boldsymbol{\Omega}^{i}_{\alpha 1}(z), \quad (i=A, B) \end{aligned} \tag{3·45}$$

where $\boldsymbol{R}_0$ is set equal to the zero vector (i.e., $\langle\boldsymbol{k}|0\rangle=1$). Equation (3·14) is reduced to

$$\sum_{i} X^{i}(\boldsymbol{k}:z)=1. \tag{3·46}$$

Equation (3·44) together with Eqs. (3·39), (3·42) and (3·45) allows us to evaluate within SCSSA the Fourier transforms of the averaged partial propagators with which the partial spectral densities can be evaluated.

The ν-dimensional matrices $\boldsymbol{F}(z)$ and $\boldsymbol{F}^{i}(z)$ $(i=A, B)$ can be block-diagonalized by the symmetry consideration as shown in I: They can be transformed, by choosing the basis vectors of $\mathfrak{S}_n$ suitably, simultaneously into block-diagonalized matrices each block of which is associated with an irreducible representation contained in the reduction of $\mathfrak{S}_n$ into irreducible representations of the point group for site n in the "averaged" system. The same is true for $\boldsymbol{\Omega}^{i}(z)$ $(i=A, B)$. Now, noting that the one-dimensional space spanned by $|n\rangle$ provides an irreducible representation of the point group, we may replace the ν-dimensional matrices in

Eqs. (3·42) and (3·45) by their sub-matrices associated with the one-dimensional irreducible representation. If the quasi-particle has multiple degrees of freedom per site, e.g., a Frenkel exciton in mixed alkali-halides and a phonon, a subspace spanned by all the state vectors localized on site n provides a multi-dimensional representation of the point group of the site. The representation may be either an irreducible representation or a reducible one which can be reduced to a number of irreducible representations. Then sub-matrices of the ν-dimensional matrices in Eqs. (3·42) and (3·45) can be dropped provided that they are associated with other irreducible representations than those. This result is consistent with the fact that a quantum of external origin can excite, when absorbed or scattered at a site, only those modes with the same transformation property with respect to the point group for the site as the states localized on the site. (See Eq. (3·10))

From the time-reversal invariance, the inversion symmetry and the equation $\langle \boldsymbol{k}|0\rangle = 1$, we can conclude that $\langle 0, \alpha|\boldsymbol{k}\rangle$ is real and an even function of $\boldsymbol{k}$ in case that $|0, \alpha\rangle$ has the same transformation property as $|0, 1\rangle = |0\rangle$ with respect to the point group for site 0, i.e., $|0, \alpha\rangle\langle 0, 1|$ is invariant. Then we have the equality $\langle 0, \alpha|\boldsymbol{k}\rangle = \langle \boldsymbol{k}|0, \alpha\rangle$ which verifies that the Fourier transforms of $X^i_{\mathrm{op}}(z)$ $(i = A, B)$ are equal to those of their transposes. Note that $X^i(\boldsymbol{k}:z)$ $(i = A, B)$ are even functions of $\boldsymbol{k}$.

$G^{ij}(\boldsymbol{k}:z)$ $(i, j = A, B)$ given in SCSSA by Eqs. (3·44) retain several properties of the exact ones. Firstly, they are even functions of $\boldsymbol{k}$. Secondly, they are symmetric with respect to interchange of the superscripts i and j. This follows from the fact that $S^{ij}(z)$ $(i, j = A, B)$ given by Eqs. (3·42) have a similar symmetry on account of that $\boldsymbol{F}(z)$ and $\boldsymbol{F}^i(z)$ $(i = A, B)$ are symmetric matrices. Thirdly, they satisfy the equation of the Fourier transformed form of Eqs. (3·6) on account of Eq. (3·46) and the equation $\sum_{i,j} S^{ij}(z) = 0$, which follows from Eqs. (2·13) and (3·42).

Various results obtained so far are applicable to multi-component mixed crystals as well. We derive here for later convenience a few results which are valid only for binary mixed crystals. By making use of Eq. (2·13), Eqs. (3·42) can be written as

$$S^{ij}(z) = (2\delta_{ij} - 1)S(z), \quad (i, j = A, B) \tag{3·47}$$

$$S(z) = xy\{\boldsymbol{F}^A(z)\boldsymbol{F}(z)^{-1}\boldsymbol{F}^B(z)\}_{1,1}. \tag{3·48}$$

On the other hand, Eq. (3·38) shows that $\boldsymbol{\mathcal{Q}}^i(z)$ $(i = A, B)$ satisfy the equation

$$\boldsymbol{\mathcal{Q}}^i(z) = \mathbf{1} - \boldsymbol{\mathcal{Q}}^i(z)(\boldsymbol{\mathcal{D}}^i(z) - \boldsymbol{D}(z))\boldsymbol{F}(z). \quad (i = A, B) \tag{3·49}$$

These equations together with Eq. (3·40) yield

$$\sum_i c^i \boldsymbol{\mathcal{Q}}^i(z)(\boldsymbol{\mathcal{D}}^i(z) - \boldsymbol{D}(z)) = 0. \tag{3·50}$$

From Eqs. (3·40) and (3·50) we obtain

$$\left.\begin{aligned} x\mathfrak{Q}^A(z) &= -(\mathcal{D}^B(z)-\boldsymbol{D}(z))\boldsymbol{\Delta}(z)^{-1}, \\ y\mathfrak{Q}^B(z) &= (\mathcal{D}^A(z)-\boldsymbol{D}(z))\boldsymbol{\Delta}(z)^{-1}, \end{aligned}\right\} \tag{3·51}$$

where

$$\boldsymbol{\Delta}(z)=\mathcal{D}^A(z)-\mathcal{D}^B(z). \tag{3·52}$$

By making use of Eqs. (3·39) and (3·51), Eq. (3·48) can be written as

$$\begin{aligned} S(z) &= \{x\mathfrak{Q}^A(z)^T\boldsymbol{F}(z)y\mathfrak{Q}^B(z)\}_{1,1} \\ &= -\{\boldsymbol{\Delta}(z)^{-1}(\mathcal{D}^B(z)-\boldsymbol{D}(z))\boldsymbol{F}(z)(\mathcal{D}^A(z)-\boldsymbol{D}(z))\boldsymbol{\Delta}(z)^{-1}\}_{1,1} \\ &= \{\boldsymbol{\Delta}(z)^{-1}[x\mathcal{D}^A(z)+y\mathcal{D}^B(z)-\boldsymbol{D}(z)]\boldsymbol{\Delta}(z)^{-1}\}_{1,1}, \end{aligned} \tag{3·53}$$

where the following form of the self-consistency condition[3] has been used:

$$\boldsymbol{F}(z)=-y(\mathcal{D}^A(z)-\boldsymbol{D}(z))^{-1}-x(\mathcal{D}^B(z)-\boldsymbol{D}(z))^{-1}. \tag{3·54}$$

§ 4. Discussion

A. An essential step of the formalism presented in §3 is to neglect the cross terms in the double summations of Eqs. (3·30). We examine here this approximation.

From Eqs. (3·18), (3·20), (3·31) and (3·33) we obtain

$$G^{ij}(z)=\sum_{n\neq n'}X^i(n:z)^TG(z)X^j(n':z)+\sum_n|n\rangle\delta_{ij}c^iF^i(z)\langle n|. \quad (i,j=A,B) \tag{4·1}$$

On the other hand, the conditionally averaged propagators are related to the single-site T-matrices $t^i(n:z)$ $(i=A,B)$ by the equation

$$\mathcal{G}^i(n:z)=G(z)+G(z)t^i(n:z)G(z). \quad (i=A,B) \tag{4·2}$$

Here $t^i(n:z)$ describes the scattering of the quasi-particle due to a real atom replacing a fictitious average atom occupying site n of the average medium in SCSSA. With Eqs. (3·22) and (4·2), Eq. (3·21) is written as

$$X^i(n:z)=|n\rangle c^i\langle n|+t^i(n:z)G(z)|n\rangle c^i\langle n|. \quad (\mathrm{i=A,B}) \tag{4·3}$$

With Eqs. (3·34), (4·2) and (4·3), Eq. (4·1) is written as

$$\begin{aligned} G^{ij}=\sum_{n\neq n'}|n\rangle c^i\langle n|\{G+Gt^i(n)G+Gt^j(n')G+Gt^i(n)Gt^j(n')G\}|n'\rangle c^j\langle n'| \\ +\delta_{ij}\sum_n|n\rangle c^i\langle n|\{G+Gt^i(n)G\}|n\rangle\langle n|, \quad (i,j=A,B) \end{aligned} \tag{4·4}$$

where the argument z is suppressed. These equations can be represented diagrammatically as shown in Fig. 1. Inspecting Fig. 1 we find that all the diagrams are so-called no-crossed-line diagrams. Indeed the averaged partial propagators given by Eqs. (4·4) are exactly equal to the results to be obtained by performing the following three steps of procedures: i) χ^i_{op} and χ^j_{op} in Eq. (3·1) are expanded into their single-site counterparts $\chi^i(n)$ and $\chi^j(n)$ and also $\mathcal{G}(z)$ in the equations is

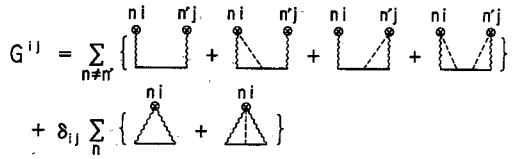

Fig. 1. The diagram representation of Eq. (4·4) determining the averaged partial propagators $G^{ij}(z)$ $(i, j=A, B)$ in terms of the single-site T-matrices $t^i(n:z)$ $(i=A, B)$. The rules by which the diagrams are drawn may be obvious. For detail, see I.

expanded in a perturbation series with respect to the single-site T-matrices.[8] ii) Configurational averaging of the resulting perturbation series for the partial propagators is performed. Here many terms of the series vanish upon averaging owing to the fact that the single-site T-matrices satisfy the self-consistency condition, i.e., they vanish when averaged. iii) Each term of the resulting perturbation series for the averaged partial propagators is represented by an appropriate diagram and it is retained in the perturbation series or dropped from this according to whether the diagram is a no-crossed-line diagram or not, respectively.

Thus it is confirmed that the approximation is consistent with the single-site approximation. Bearing in mind the diagrammatic interpretation of the approximation we can show by a similar argument as in I that the errors in the partial spectral densities evaluated by the present formalism are $O(x^2y^2/Z^2)$ or $O(x^2y^2/Z)$ according to whether the inverse of the propagator is a canonical random operator or not, respectively, where Z is the coordination number of the lattice.

Recently Harris et al.[9] evaluated within SCSSA the configurational average of the host-host Green's function (a partial propagator) of a magnon in a diluted Heisenberg ferromagnet by making use of a different diagrammatic method, i.e., Leath's method.[10] Their result can be shown to be identical with the result to be obtained if the present formalism is applied to this particular problem.

Incidentally we remark that the diagonal elements of $G^{ij}(z)$ $(i, j=A, B)$ in the site-representation are equal to $\delta_{ij}c^iF^i(z)$ $(i, j=A, B)$, respectively, as easily found in Eqs. (4·1) and (3·21). These results are, however, general relations and can be verified directly from the definitions of the relevant quantities with no recourse to SCSSA.

B. We compare here the present formalism with others. If the inverse of the propagator is a diagonal random operator, various ν-dimensional matrices appeared in previous sections are reduced to ordinary numbers since $\mathfrak{S}_n$ is then a one-dimensional space spanned by $|n\rangle$. We denote them by the same but non-bold-faced letters as before. $X^i(n:z)$ $(i=A, B)$ are localized on site n; $X^i(n:z) = |n\rangle X^i(z)\langle n|$ $(i=A, B)$, where $X^i(z) \equiv c^i\Omega^i(z)$ $(=c^iF^i(z)/F(z))$ $(i=A, B)$, which satisfy $\sum_i X^i(z)=1$. Therefore we obtain

$$G^{ij}(\boldsymbol{k}:z) = X^i(z)X^j(z)G(\boldsymbol{k}:z) + \{\delta_{ij}X^i(z) - X^i(z)X^j(z)\}F(z). \qquad (i, j=A, B) \qquad (4\cdot5)$$

Here $G(\boldsymbol{k}:z)$ in CPA takes the form

$$G(\boldsymbol{k}:z)=(D(z)-\varepsilon_0(\boldsymbol{k}))^{-1}. \tag{4·6}$$

As will be shown in Appendix I, Eqs. (4·5) are equivalent to the results to be obtained by using the method of Elliott et al. together with CPA.

We can show that the present formalism leads to the same results for the averaged partial propagators as BEB within the common range of applicability, i.e., for the case where the inverse of the propagator or its renormalized form is not only an additive random operator but also a canonical one. This will be described in Appendix II.

C. Strictly speaking, SPEF can be related to the averaged partial propagators only if the coupling constant between a quasi-particle and a quantum of external origin depends only on the type of the atom occupying the site where they interact with each other. This limitation prevents the present formalism from applying to the coherent one-phonon inelastic neutron scattering intensity*) of a substitutionally disordered lattice, say, a binary mixed crystal A_xB_y since the neutron scattering lengths of the nuclei are modified by the Debye-Waller factors each of which is a random variable depending not only on the type of the nucleus occupying the relevant site but also on those of other nuclei occupying near-by sites.[7),11)] Fortunately, as explained in Appendix III, we can approximate the Debye-Waller factors so that the present formalism is applicable to this problem. Here it is very important that the approximation is consistent with SCSSA.

The incoherent one-phonon inelastic neutron scattering intensity contains only the diagonal elements of the partial propagators of a phonon but not the off-diagonal ones.[7),11)] Therefore if the Debye-Waller factors modifying the incoherent neutron scattering lengths of the nuclei are replaced by their approximate expressions given in Appendix III we can evaluate also the incoherent intensity within SCSSA since the diagonal elements of the averaged partial propagators are known as remarked at the end of part A of the present section.

Kaplan and Mostoller investigated recently the lattice vibration of mixed crystal $(NH_4)_xK_yCl$ by taking into account the force constant changes.[4)] Their method of evaluating the averaged phonon propagator is the same as that proposed in I but their treatment of the coherent neutron scattering intensity is less accurate than that presented in this paper.

Incidentally we remark that the approximate expressions for the Debye-Waller factors mentioned above are good also for the coherent elastic neutron intensity.

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*) The one-phonon inelastic neutron scattering intensity of a lattice consists of the two parts, i.e., the coherent part and the incoherent part with respect to random orientations of the nuclear spins.[11)]

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Appendix I

Equations (3·51) yield different expressions for $X^i(z)$ $(i=A, B)$ from those presented in the text:

$$X^A(z)=-(\mathcal{D}^B(z)-D(z))/\Delta(z), \quad X^B(z)=(\mathcal{D}^A(z)-D(z))/\Delta(z), \quad \text{(AI·1)}$$

where $\Delta(z)\equiv\mathcal{D}^A(z)-\mathcal{D}^B(z)$. On the other hand, by making use of Eqs. (3·47) and (3·53), the second terms of Eq. (4·5) can be written in different forms:

$$G^{ij}(\boldsymbol{k}:z)=X^i(z)X^j(z)G(\boldsymbol{k}:z)$$
$$+(2\delta_{ij}-1)\{x\mathcal{D}^A(z)+y\mathcal{D}^B(z)-D(z)\}/\Delta(z)^2. \quad (i,j=A,B) \quad \text{(AI·2)}$$

Equations (AI·2) with Eqs. (AI·1) are the same results as those to be obtained if the method of Elliott et al.[8] is applied to the propagator $\mathcal{G}(z)=(\sum_n|n\rangle\mathcal{D}_n(z)\cdot\langle n|-H_0)^{-1}$. (cf., Eqs. (2·140)$\sim$(2·142) in Ref. 6), which are derived by using the method together with CPA.)

Appendix II

For simplicity, we limit our argument, as in I, to the special case where the inverse of the propagator

$$\mathcal{G}(z)^{-1}=\sum_n|n\rangle\mathcal{D}_n(z)\langle n|-\sum_{n\neq n'}|n\rangle h_{nn'}\langle n'| \quad \text{(AII·1)}$$

is a random operator such that diagonal element $\mathcal{D}_n(z)$ is a random variable taking one of the two possible values $\mathcal{D}^i(z)=\eta^i(z-\varepsilon^i)$ $(i=A, B)$ and off-diagonal element $h_{nn'}$ is a random variable of the form $h_{nn'}=(1/2)(W_n+W_{n'})h^0_{nn'}$. Here $h^0_{nn'}$ is the transfer integral of a periodic Hamiltonian and $W_n(W_{n'})$ is a random variable taking W^A or W^B depending on the type of the atom on $n(n')$.

$\mathfrak{S}_n$ is a two-dimensional space spanned by

$$|n,1\rangle=|n\rangle, \quad |n,2\rangle=\sum_{n'(\neq n)}h^0_{n'n}|n'\rangle. \quad \text{(AII·2)}$$

Hence we have

$$\mathcal{D}^i(z)=\begin{pmatrix}\mathcal{D}^i(z) & -W^i/2\\ -W^i/2 & 0\end{pmatrix}, \quad (i=A,B) \quad \text{(AII·3)}$$

where a quantity denoted by H_0 in the general formalism in § 2 is set equal to zero. If we parametrize $\boldsymbol{D}(z)$ as

$$\boldsymbol{D}(z)=\begin{pmatrix}D(z) & -W(z)/2\\ -W(z)/2 & -V(z)\end{pmatrix}, \quad \text{(AII·4)}$$

we obtain

$$G(\boldsymbol{k}:z)=\{D(z)-W(z)\varepsilon_0(\boldsymbol{k})-V(z)\varepsilon_0(\boldsymbol{k})^2\}^{-1}, \tag{AII·5}$$

where

$$\varepsilon_0(\boldsymbol{k})=\sum_{n'(\neq n)} h^0_{n'n}\exp\{i\boldsymbol{k}\cdot(\boldsymbol{R}_n-\boldsymbol{R}_{n'})\}. \tag{AII·6}$$

Equations (3·45), (3·51), (3·52), (AII·2) (AII·3), (AII·4) and (AII·6) yield

$$\left.\begin{aligned} X^A(\boldsymbol{k}:z)&=-\{(W^B-W(z))/2-V(z)\varepsilon_0(\boldsymbol{k})\}/\kappa,\\ X^B(\boldsymbol{k}:z)&=\{(W^A-W(z))/2-V(z)\varepsilon_0(\boldsymbol{k})\}/\kappa,\end{aligned}\right\} \tag{AII·7}$$

where $\kappa=(W^A-W^B)/2$. On the other hand, Eqs. (3·52), (3·53), (AII·3) and (AII·4) yield

$$S(z)=V(z)/\kappa^2. \tag{AII·8}$$

Equations (3·44), (3·47), (AII·7) and (AII·8) together with an elementary manipulation yield

$$\boldsymbol{G}(\boldsymbol{k}:z)\equiv\begin{pmatrix}G^{AA}(\boldsymbol{k}:z) & G^{AB}(\boldsymbol{k}:z)\\ G^{BA}(\boldsymbol{k}:z) & G^{BB}(\boldsymbol{k}:z)\end{pmatrix}=\begin{pmatrix}R^{AA}(\boldsymbol{k}:z) & R^{AB}(\boldsymbol{k}:z)\\ R^{BA}(\boldsymbol{k}:z) & R^{BB}(\boldsymbol{k}:z)\end{pmatrix}^{-1}, \tag{AII·9}$$

where

$$\begin{aligned}R^{ij}(\boldsymbol{k}:z)=&D(z)+(W^i-W(z))(W^j-W(z))/4V(z)\\&-(1/2)(W^i+W^j)\varepsilon_0(\boldsymbol{k})\quad(i,j=A,B)\end{aligned} \tag{AII·10}$$

and use has been made of the equation

$$\det(R^{ij}(\boldsymbol{k}:z))=\kappa^2\{D(z)-W(z)\varepsilon_0(\boldsymbol{k})-V(z)\varepsilon_0(\boldsymbol{k})^2\}/V(z). \tag{AII·11}$$

Now the partial propagator matrix $\boldsymbol{G}(\boldsymbol{k}:z)$ derived by making use of BEB is given by Eq. (AIII·16) in I. Noting Eqs. (AIII·21) in I which relate the parameters in BEB with those in the present formalism we see that both formalisms present identical results for the Fourier transforms of the averaged partial propagators.

Appendix III

——*The Self-Consistent Single-Site Approximation for the Debye-Waller Factors*——

The coherent neutron scattering length a_n of the nucleus occupying site n of a binary mixed crystal A_xB_y enters in the coherent one-phonon inelastic neutron scattering intensity through the modified form:

$$\tilde{a}_n=a_n\exp\{-W_n(\boldsymbol{q}:T)\}, \tag{AIII·1}$$

where $W_n(\boldsymbol{q}:T)$ is given by the thermal average of $(\boldsymbol{q}\cdot\boldsymbol{u}_n)^2/2$ with $\boldsymbol{u}_n$ being the

displacement vector of the nucleus on n relative to its equilibrium position. After an elementary manipulation (see Eq. (II·62) in Ref. 11)), we obtain

$$W_n(\boldsymbol{q}:T)=\hbar\int_0^\infty d\omega\{n_T(\hbar\omega)+\tfrac{1}{2}\}\sum_{\alpha\beta=1}^{3}q_\alpha q_\beta(-\pi^{-1})\,\mathrm{Im}\,\mathcal{G}_{n\alpha,n\beta}(\omega^2+i0), \quad \text{(AIII·2)}$$

where $n_T(\hbar\omega)\equiv\{\exp(\hbar\omega/k_BT)-1\}^{-1}$ and $\mathcal{G}(\omega^2+iO)\equiv(\mathcal{G}_{n\alpha,n'\beta}(\omega^2+i0))$ is the propagator (matrix) of a phonon, i.e., the Fourier transform with respect to the time variable of the retarded displacement-displacement correlation.

If $W_n(\boldsymbol{q}:T)$ is approximated by a random variable depending only on the type of the nucleus occupying site n, $\tilde{a}_n$ becomes to a similar random variable, which allows us to evaluate the neutron intensity by making use of the formalism presented in the text. A natural approximation is to replace $W_n(\boldsymbol{q}:T)$ by its conditional average $\hat{W}_n(\boldsymbol{q}:T)$ which is a random variable taking one of the two possible values:

$$\hat{W}^i(\boldsymbol{q}:T)=\langle W_n(\boldsymbol{q}:T)\rangle^{i/n}. \quad (i=A,B) \qquad \text{(AIII·3)}$$

Let $\overleftrightarrow{F}^i(z)\equiv(F^i_{\alpha\beta}(z))$ $(i=A,B)$ be second rank tensors defined in terms of the conditionally averaged propagators $\mathcal{G}^i(n:z)$ $(i=A,B)$ by the equations: $F^i_{\alpha\beta}(z)\equiv\mathcal{G}^i_{n\alpha,n\beta}(n:z)$ $(i=A,B;\ \alpha,\beta=1,2,3)$. Then Eqs. (AIII·2) and (AIII·3) yield

$$\hat{W}^i(\boldsymbol{q}:T)=\int_0^\infty d\omega^2\{\hbar\boldsymbol{q}\cdot\overleftrightarrow{\rho}^i(\omega^2)\cdot\hbar\boldsymbol{q}/2M^i\}\,(1/2\hbar\omega)\coth(\hbar\omega/2k_BT), \quad (i=A,B) \qquad \text{(AIII·4)}$$

$$\overleftrightarrow{\rho}^i(\omega^2)=M^i(-\pi^{-1})\,\mathrm{Im}\,\overleftrightarrow{F}^i(\omega^2+i0), \quad (i=A,B) \qquad \text{(AIII·5)}$$

where M^A and M^B are the masses of two types of nuclei. $\overleftrightarrow{\rho}^i(\omega^2)$ $(i=A,B)$ are symmetric tensors normalized as

$$\int_0^\infty d\omega^2\overleftrightarrow{\rho}^i(\omega^2)=\overleftrightarrow{1}. \quad (i=A,B) \qquad \text{(AIII·6)}$$

Here $\overleftrightarrow{1}\equiv(\delta_{\alpha\beta})$ is the unit tensor. $\tilde{a}_n$ takes now one of the two possible values of the modified neutron scattering lengths:

$$\tilde{a}^i=a^i\exp\{-\hat{W}^i(\boldsymbol{q}:T)\}, \quad (i=A,B) \qquad \text{(AIII·7)}$$

where a^A and a^B are two neutron scattering lengths before modified.

We can prove that the above approximation for the Debye-Waller factors is consistent with SCSSA and that an error brought in the neutron intensity by the approximation is $O(x^2y^2/Z)$ or $O(x^2y^2/Z^2)$ according to whether the force constants change or not, respectively. The proof is rather lengthy and shall not be given here.

If the lattice has a cubic symmetry, $\overleftrightarrow{\rho}^i(\omega^2)$ $(i=A,B)$ are reduced to the scalars $\rho^i(\omega^2)$ $(i=A,B)$ multiplied by $\overleftrightarrow{1}$. Then the quantities in the brackets of Eq. (AIII·4) are written as $T^i(\hbar q)\rho^i(\omega^2)$ $(i=A,B)$, where $T^i(\hbar q)\equiv(\hbar q)^2/2M^i$ $(i=A,B)$ are the recoil energies of the two types of nuclei. Note that

$\rho^i(\omega^2)$ $(i=A, B)$ are nothing but the partial densities of states of a phonon per site, one polarization and unit squared frequency.

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