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# Electronic structure and response functions in random alloys: an application of block recursion and Green matrices

Kamal Krishna Saha and Abhijit Mookerjee

S N Bose National Centre for Basic Sciences, Block-JD, Sector-III, Salt Lake City, Kolkata-700098, India

E-mail: kamal@bose.res.in

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## Abstract

We present here a generalization of the recursion method of Haydock *et al* (1972 *J. Phys. C: Solid State Phys.* **5** 2845) for the calculation of Green matrices (in angular momentum space). Earlier approaches concentrated on the diagonal elements, since the focus was on spectral densities. However, calculations of configuration averaged response functions or neutron scattering cross-sections require the entire Green matrices and self-energy matrices obtained from them. This necessitated the generalization of the recursion method presented here, with examples.

## 1. Introduction

Augmented space recursion carried out in a minimal basis set representation of the tight-binding linear muffin-tin orbitals method (TB-LMTO-ASR) has been proposed earlier by us [2, 3] as a technique for the incorporation of the effects of configuration fluctuations for random substitutionally disordered alloys. This can be achieved without the usual problems of violation of the Herglotz analytic properties<sup>1</sup> of the approximated configuration averaged Green functions for the Schrödinger equation for these random alloys. Although our initial focus was on configuration averages of the density of states and spectral functions, recently we have proposed using the TB-LMTO-ASR for the study of configuration averaged optical conductivities [4] or coherent and incoherent neutron scattering cross-sections [5]. These calculations require the full Green matrices in angular momentum space and not only their diagonal elements. We propose here the use of a generalization of the recursion method of Haydock *et al* [1]. The block recursion technique had been introduced earlier by Godin and Haydock [6, 7] in the very different context for obtaining the scattering  $S$ -matrix for finite

<sup>1</sup> A function of a complex variable  $f(z)$  is called Herglotz if: all singularities of  $f(z)$  lie on the real  $z$  axis and  $\text{Im } f(z) = -\text{sgn}(\text{Im } z)$  for all  $z$  off the real axis.

scatterers attached to perfect leads. The block recursion has been discussed in a general context by Nex [8] and Haydock *et al* [9]. Earlier, Inoue and Ohta [10] had proposed the use of an orbital symmetrized version of the block recursion method for electronic structure calculations. We shall borrow their ideas and set up a block recursion in angular momentum space (rather than the lead space, as in Godin and Haydock's work) in order to obtain the Green matrices (in angular momentum space) directly. Unlike in the earlier works, the application will be to configuration averages in random alloys.

## 2. Methodology

### 2.1. The TB-LMTO Hamiltonian in augmented space and the recursion method

The augmented space recursion based on the tight-binding linear muffin-tin orbitals method (TB-LMTO-ASR) has been described thoroughly in a series of articles [11–19]. We shall introduce the salient features of the ASR which will be required by us in our subsequent discussions.

We shall start from first-principles tight-binding linear muffin-tin orbitals (TB-LMTO) [20, 21] in the most localized representation ( $\alpha$  representation). This is necessary, because the subsequent recursion requires a sparse representation of the Hamiltonian. In this representation, the second-order alloy Hamiltonian is given by

$$\mathbf{H}^{(2)} = \mathbf{E}_v + \mathbf{h} - \mathbf{h}\mathbf{o}\mathbf{h}$$

where

$$\begin{aligned} \mathbf{h} &= \sum_R (\mathbf{C}_R - \mathbf{E}_{vR}) \mathcal{P}_R + \sum_R \sum_{R'} \Delta_R^{1/2} \mathbf{S}_{RR'} \Delta_{R'}^{1/2} \mathcal{T}_{RR'} \\ \mathbf{o} &= \sum_R \mathbf{o}_R \mathcal{P}_R \end{aligned} \quad (1)$$

$\mathbf{C}_R$ ,  $\mathbf{E}_{vR}$ ,  $\Delta_R$  and  $\mathbf{o}_R$  are diagonal matrices in angular momentum space:

$$\begin{aligned} \mathbf{C}_R &= C_{RL} \delta_{LL'} & \mathbf{E}_{vR} &= E_{vRL} \delta_{LL'} \\ \Delta_R &= \Delta_{RL} \delta_{LL'} & \text{and} & \quad \mathbf{o}_R = o_{RL} \delta_{LL'} \end{aligned}$$

and  $\mathbf{S}_{RR'} = S_{RL,R'L'}$  is a matrix of rank  $L_{\max}$ .  $\mathcal{P}_R = |R\rangle\langle R|$  and  $\mathcal{T}_{RR'} = |R\rangle\langle R'|$  are projection and transfer operators in the Hilbert space  $\mathcal{H}$  spanned by the tight-binding basis  $\{|R\rangle\}$ . Here,  $R$  relates to the position of atoms in the solid and  $L$  is a composite label  $\{\ell, m, m_s\}$  for the angular momentum quantum numbers.  $\mathbf{C}$ ,  $\Delta$  and the  $\mathbf{o}$ 's are potential parameters of the TB-LMTO method; these are diagonal matrices in the angular momentum indices,  $\mathbf{o}^{-1}$  has the dimension of energy and the  $\mathbf{E}_v$ 's are the energy windows about which the muffin-tin orbitals are linearized.

For a disordered binary alloy we may write

$$\begin{aligned} C_{RL} &= C_L^A n_R + C_L^B (1 - n_R) \\ \Delta_{RL}^{1/2} &= (\Delta_L^A)^{1/2} n_R + (\Delta_L^B)^{1/2} (1 - n_R) \\ o_{RL} &= o_L^A n_R + o_L^B (1 - n_R). \end{aligned} \quad (2)$$

Here  $\{n_R\}$  are the random site-occupation variables which take values 1 and 0 depending upon whether the muffin tin labelled by  $R$  is occupied by an A or B type of atom. The atom sitting at  $\{R\}$  can be either of type A ( $n_R = 1$ ) with probability  $x$  or of type B ( $n_R = 0$ ) with probability  $y$ . The augmented space formalism (ASF) now introduces the space of configurations of the set of binary random variables  $\{n_R\} : \Phi$ .

In the absence of short ranged order, each random variable  $n_R$  has associated with it an operator  $\mathbf{M}_R$  whose spectral density is its probability density:

$$\begin{aligned} p(n_R) &= x\delta(n_R - 1) + y\delta(n_R) \\ &= -\frac{1}{\pi} \lim_{\delta \rightarrow 0} \text{Im} \langle \uparrow_R | [(n_R + i\delta)\mathbf{I} - \mathbf{M}_R]^{-1} | \uparrow_R \rangle \end{aligned} \quad (3)$$

where  $\mathbf{M}_R$  is an operator whose eigenvalues 1, 0 correspond to the observed values of  $n_R$  and whose corresponding eigenvectors  $\{|1_R\rangle, |0_R\rangle\}$  span a configuration space  $\phi_R$  of rank 2. We may change the basis to  $\{|\uparrow_R\rangle, |\downarrow_R\rangle\}$ :

$$\begin{aligned} |\uparrow_R\rangle &= \sqrt{x}|0_R\rangle + \sqrt{y}|1_R\rangle \\ |\downarrow_R\rangle &= \sqrt{y}|0_R\rangle - \sqrt{x}|1_R\rangle \end{aligned}$$

and in this new basis the operator is

$$n_R \rightarrow \mathbf{M}_R = x\mathcal{P}_R^\uparrow + y\mathcal{P}_R^\downarrow + \sqrt{xy}(\mathcal{T}_R^{\uparrow\downarrow} + \mathcal{T}_R^{\downarrow\uparrow}). \quad (4)$$

These two vectors span the space  $\phi_R$ . The full configuration space  $\Phi = \prod_R^\otimes \phi_R$  is then spanned by vectors of the form  $|\uparrow \uparrow \downarrow \uparrow \downarrow \dots\rangle$ . These configurations may be labelled by the sequence of sites  $\{\mathcal{C}\}$  at which we have a  $\downarrow$ . For example, for the state just quoted  $\{\mathcal{C}\} = \{3, 5, \dots\}$ . This sequence is called the *cardinality sequence*. If we define the configuration  $|\uparrow \uparrow \dots \uparrow \dots\rangle$  as the *average* or *reference* configuration, then the *cardinality sequence* of the *reference* configuration is the null sequence  $\{\emptyset\}$ .

The augmented space theorem [11] states that

$$\langle\langle \mathbf{A}(\{n_R\}) \rangle\rangle = \langle\langle \emptyset | \tilde{\mathbf{A}} | \emptyset \rangle\rangle \quad (5)$$

where

$$\tilde{\mathbf{A}}(\{\mathbf{M}_R\}) = \int \dots \int \mathbf{A}(\{\lambda_R\}) \prod d\mathbf{P}(\lambda_R).$$

$\mathbf{P}(\lambda_R)$  is the spectral density of the self-adjoint operator  $\mathbf{M}_R$ .

Applying this idea we may obtain exact expressions for configuration averages of Green matrices (in angular momentum space) both in the real and reciprocal space representations:

$$\langle\langle \mathbf{G}(R, R, z) \rangle\rangle = \langle R \otimes \{\emptyset\} | (z\tilde{\mathbf{I}} - \tilde{\mathbf{H}}^{(2)})^{-1} | R \otimes \{\emptyset\} \rangle \quad (6)$$

$$\langle\langle \mathbf{G}(\mathbf{k}, z) \rangle\rangle = \langle \mathbf{k} \otimes \{\emptyset\} | (z\tilde{\mathbf{I}} - \tilde{\mathbf{H}}^{(2)})^{-1} | \mathbf{k} \otimes \{\emptyset\} \rangle \quad (7)$$

where  $\mathbf{G}$  and  $\mathbf{H}^{(2)}$  are operators which are matrices in angular momentum space, and the augmented  $k$ -space basis  $|\mathbf{k}, L \otimes \{\emptyset\}\rangle$  has the form

$$(1/\sqrt{N}) \sum_R \exp(-i\mathbf{k} \cdot R) |R, L \otimes \{\emptyset\}\rangle.$$

The augmented space Hamiltonian  $\tilde{\mathbf{H}}^{(2)}$  is constructed from the TB-LMTO Hamiltonian  $\mathbf{H}^{(2)}$  by replacing each random variable  $n_R$  by operators  $\mathbf{M}_R$ . It is an operator in the augmented space  $\Psi = \mathcal{H} \otimes \Phi$ . The ASF maps a disordered Hamiltonian described in a Hilbert space  $\mathcal{H}$  onto an ordered Hamiltonian in an enlarged space  $\Psi$ , where the space  $\Psi$  is constructed as the outer product of the space  $\mathcal{H}$  and configuration space  $\Phi$  of the random variables of the disordered Hamiltonian. The configuration space  $\Phi$  is of rank  $2^N$  if there are  $N$  muffin-tin spheres in the system. Another way of looking at  $\tilde{\mathbf{H}}^{(2)}$  is to note that it is the *collection* of all possible Hamiltonians for all possible configurations of the system.

A little mathematics yields the following:

$$\langle\langle \mathbf{G}(z) \rangle\rangle = \langle \mathbf{1} | (\tilde{\mathbf{A}} + \tilde{\mathbf{B}} + \tilde{\mathbf{F}} - \tilde{\mathbf{S}} + (\tilde{\mathbf{J}} + \tilde{\mathbf{S}})\tilde{\mathbf{o}}(\tilde{\mathbf{J}} + \tilde{\mathbf{S}}))^{-1} | \mathbf{1} \rangle. \quad (8)$$

For real space calculations,

$$|\mathbf{1}\rangle = \frac{\mathbf{A}(\Delta^{-1/2})}{[\mathbf{A}(\Delta^{-1})]^{1/2}} |R \otimes \{\emptyset\}\rangle + \frac{\mathbf{F}(\Delta^{-1/2})}{[\mathbf{A}(\Delta^{-1})]^{1/2}} |R \otimes \{R\}\rangle,$$

and for reciprocal space calculations,

$$|\hat{\mathbf{1}}\rangle = \frac{\mathbf{A}(\Delta^{-1/2})}{[\mathbf{A}(\Delta^{-1})]^{1/2}} |\mathbf{k} \otimes \{\emptyset\}\rangle + \frac{\mathbf{F}(\Delta^{-1/2})}{[\mathbf{A}(\Delta^{-1})]^{1/2}} |\mathbf{k} \otimes \{R\}\rangle,$$

where

$$\tilde{\mathbf{K}} = \sum_R \{ \mathbf{K} ((z\mathbf{I} - \mathbf{C})\Delta^{-1}) / \mathbf{A}(\Delta^{-1}) \} \mathcal{O}_K$$

and  $\mathbf{K}$  may be  $\mathbf{A}$ ,  $\mathbf{B}$  or  $\mathbf{F}$ , while the operators

$$\mathcal{O}_A = \mathcal{P}_R \otimes \mathcal{I} \quad \mathcal{O}_B = \mathcal{P}_R \otimes \mathcal{P}_R^\downarrow \quad \mathcal{O}_F = \mathcal{P}_R \otimes \{ \mathcal{T}_R^{\uparrow\downarrow} + \mathcal{T}_R^{\downarrow\uparrow} \}.$$

Moreover,  $\tilde{\mathbf{J}} = \tilde{\mathbf{J}}_A + \tilde{\mathbf{J}}_B + \tilde{\mathbf{J}}_F$  and  $\tilde{\mathbf{o}} = \tilde{\mathbf{o}}_A + \tilde{\mathbf{o}}_B + \tilde{\mathbf{o}}_F$  where

$$\begin{aligned} \tilde{\mathbf{J}}_K &= \sum_R \{ \mathbf{K} ((\mathbf{C} - \mathbf{E}_v)\Delta^{-1}) / \mathbf{A}(\Delta^{-1}) \} \mathcal{O}_K \\ \tilde{\mathbf{o}}_K &= \sum_R \{ \mathbf{K}(\mathbf{o}\Delta)\mathbf{A}(\Delta^{-1}) \} \mathcal{O}_K. \end{aligned} \quad (9)$$

For any diagonal (in angular momentum space) operator  $\mathbf{V}$ ,

$$\begin{aligned} \mathbf{A}(\mathbf{V}) &= (xV_L^A + yV_L^B)\delta_{LL'}, \\ \mathbf{B}(\mathbf{V}) &= (y-x)(V_L^A - V_L^B)\delta_{LL'}, \\ \mathbf{F}(\mathbf{V}) &= \sqrt{xy}(V_L^A - V_L^B)\delta_{LL'}. \end{aligned}$$

In the case where is no off-diagonal disorder due to local lattice distortion because of size mismatch,

$$\tilde{\mathbf{S}} = \sum_R \sum_{R'} \mathbf{A}(\Delta^{-1})^{-1/2} \mathbf{S}_{RR'} \mathbf{A}(\Delta^{-1})^{-1/2} \mathcal{T}_{RR'} \otimes \mathcal{I}.$$

Equation (8) is now exactly in the form for which the recursion method may be applied. For ordinary recursion,  $|\mathbf{1}\rangle$  is labelled by a particular  $L$  and has a representation as a  $1 \times N_{\max}$  column vector, with  $N_{\max}$  being the size in augmented space. But in the case of block recursion, we deal with an  $L \times L'$  matrix and the length of  $|\mathbf{1}\rangle$  is  $L \times L' \times N_{\max}$ . We should note that at this point the above expression for the averaged  $\langle\langle G_{RL,RL'}(z) \rangle\rangle$  or  $\langle\langle G_{LL'}(\mathbf{k}, z) \rangle\rangle$  is *exact*.

It is important to note that the operators  $\tilde{\mathbf{A}}$ ,  $\tilde{\mathbf{B}}$ ,  $\tilde{\mathbf{F}}$ ,  $\tilde{\mathbf{J}}_A$ ,  $\tilde{\mathbf{J}}_B$ ,  $\tilde{\mathbf{J}}_F$ ,  $\tilde{\mathbf{o}}_A$ ,  $\tilde{\mathbf{o}}_B$  and  $\tilde{\mathbf{o}}_F$  are all projection operators in real space (i.e. unit operators in  $\mathbf{k}$ -space) and act on an augmented space basis only to change the configuration part (i.e. the cardinality sequence  $\{C\}$ ):

$$\begin{aligned} \tilde{\mathbf{A}}|\{C\}\rangle &= A_1|\{C\}\rangle & \tilde{\mathbf{B}}|\{C\}\rangle &= A_2|\{C\}\rangle\delta(R \in \{C\}) & \tilde{\mathbf{F}}|\{C\}\rangle &= A_3|\{C \pm R\}\rangle \\ \tilde{\mathbf{J}}_A|\{C\}\rangle &= J_1|\{C\}\rangle & \tilde{\mathbf{J}}_B|\{C\}\rangle &= J_2|\{C\}\rangle\delta(R \in \{C\}) & \tilde{\mathbf{J}}_F|\{C\}\rangle &= J_3|\{C \pm R\}\rangle \\ \tilde{\mathbf{o}}_A|\{C\}\rangle &= o_1|\{C\}\rangle & \tilde{\mathbf{o}}_B|\{C\}\rangle &= o_2|\{C\}\rangle\delta(R \in \{C\}) & \tilde{\mathbf{o}}_F|\{C\}\rangle &= o_3|\{C \pm R\}\rangle. \end{aligned}$$

The coefficients  $A_1$ - $A_3$ ,  $J_1$ - $J_3$  and  $o_1$ - $o_3$  have been expressed in equation (9). The remaining operator  $\tilde{\mathbf{S}}$  is off-diagonal in real space, but diagonal in  $\mathbf{k}$ -space.

In the real space representation,

$$\tilde{\mathbf{S}}|R, \{C\}\rangle = \sum_{\chi} \mathbf{S}(\chi)|R + \chi, \{C\}\rangle.$$

Here the  $\chi$ 's are the nearest neighbour vectors. The operator  $\tilde{\mathbf{S}}$  shifts the real space site to its nearest neighbour position by the vector  $\chi$  and configuration space remains unchanged.

However, in the reciprocal space representation, it acts on an augmented space only to change the configuration part:

$$\tilde{\mathbf{S}}|\{C\}\rangle = \sum_{\chi} \exp(-i\mathbf{k} \cdot \chi) \mathbf{S}(\chi) |\{C - \chi\}\rangle.$$

Here the operator rigidly shifts the entire down-spin configuration by the vector  $\chi$ . The operation of the effective Hamiltonian is thus entirely in the configuration space.

As long as we wish to obtain only the diagonal elements of the Green function, as is required for the local density of states or the spectral densities, the ordinary recursion as described by Haydock *et al* [1] suffices. However, for response functions we shall need the full Green matrix. This will be described in the following section.

## 2.2. The configuration averaged current–current correlation function

We shall quote here the result for the dominant term in the configuration averaged current–current correlation function reported by us earlier [4]. This correlation function is directly related to the optical conductivity. We note that the expression involves the full Green matrix in angular momentum space and not only its diagonal elements.

The expression for the correlation function is

$$\langle\langle \mathcal{S}(z_1, z_2) \rangle\rangle = \int_{\text{BZ}} \frac{d^3\mathbf{k}}{8\pi^3} \text{Tr} [\mathbf{J}^{\text{eff}}(\mathbf{k}, z_1, z_2) \langle\langle \mathbf{G}^v(\mathbf{k}, z_1) \rangle\rangle \mathbf{J}^{\text{eff}}(\mathbf{k}, z_1, z_2)^\dagger \langle\langle \mathbf{G}^c(\mathbf{k}, z_2) \rangle\rangle] \quad (10)$$

and the renormalized current term is given by

$$\begin{aligned} \mathbf{J}^{\text{eff}}(\mathbf{k}, z_1, z_2) = & \langle\langle \mathbf{j}(\mathbf{k}) \rangle\rangle + 2 [\Sigma(\mathbf{k}, z_2) \mathbf{f}(z_2) \mathbf{j}^{(1)}(\mathbf{k}) + \mathbf{j}^{(1)}(\mathbf{k}) \mathbf{f}(z_1) \Sigma(\mathbf{k}, z_1)] \\ & + \Sigma(\mathbf{k}, z_2) \mathbf{f}(z_2) \mathbf{j}^{(2)}(\mathbf{k}) \mathbf{f}(z_1) \Sigma(\mathbf{k}, z_1) \end{aligned} \quad (11)$$

where

$$\mathbf{f}(z) = f_{LL'}(z) = \left\langle \frac{1}{\Delta_L} \right\rangle \left[ \frac{C_L^A}{\Delta_L^A} - \frac{C_L^B}{\Delta_L^B} - z \left( \frac{1}{\Delta_L^A} - \frac{1}{\Delta_L^B} \right) \right]^{-1} \delta_{LL'}$$

and

$$\Sigma = \mathbf{g}^{-1} - \mathbf{G}^{-1}.$$

$\mathbf{g}$  is the virtual crystal Green function. The interested reader is referred to an earlier work [4] which derives these expressions in some detail. The main point in setting these equations out is to note that in such calculations one needs the *full* Green matrix in angular momentum space. This is the main motivation for this work.

## 2.3. Setting up the block recursion

The first step in setting up the block recursion procedure is to systematically renumber the real space basis with integers. An example on a square lattice is shown in figure 1.

The nearest neighbour map is now generated by a systematic numbering of the states in augmented space as follows.

(A) *Real space formulation.* We start with numbering  $|R, \{\emptyset\}\rangle$  as 1, and then recursively generate the neighbours by acting on the states with  $\tilde{\mathbf{S}}$  and  $\tilde{\mathbf{F}}$ . Let us take an example of a square lattice:

- (i)  $\tilde{\mathbf{S}}$  acting on  $|1, \{\emptyset\}\rangle \equiv |1\rangle$  gives four new neighbours  $|2, \{\emptyset\}\rangle \cdots |5, \{\emptyset\}\rangle$ . The four real space neighbours of  $|1\rangle$  are then  $|2\rangle, |3\rangle, |4\rangle$  and  $|5\rangle$ .

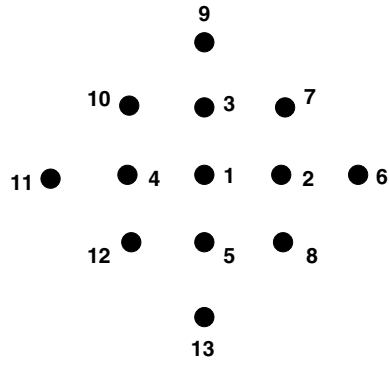


Figure 1. Systematic discrete numbering of the nearest neighbour map on a square lattice.

- (ii)  $\tilde{\mathbf{F}}$  acting on  $|1, \{\emptyset\}\rangle \equiv |1\rangle$  gives  $|1, \{1\}\rangle$ . This we number  $|6\rangle$ .
- (iii)  $\tilde{\mathbf{S}}$  acting on  $|2, \{\emptyset\}\rangle \equiv |2\rangle$  gives  $|6, \{\emptyset\}\rangle, |7, \{\emptyset\}\rangle, |1, \{\emptyset\}\rangle$  and  $|8, \{\emptyset\}\rangle$ . These we number  $|7\rangle, |8\rangle, |1\rangle$  and  $|9\rangle$ .
- (iv)  $\tilde{\mathbf{F}}$  acting on  $|2, \{\emptyset\}\rangle \equiv |2\rangle$  gives  $|2, \{2\}\rangle \equiv |10\rangle$ .

We proceed exactly as above and finally obtain the nearest neighbour map matrix, the  $n$ th column of whose  $m$ th row gives the  $n$ th neighbour of  $m$ . We show below the initial part of the nearest neighbour map for the above example:

$$\begin{pmatrix} 2 & 3 & 4 & 5 & 6 \\ 7 & 8 & 1 & 9 & 10 \\ 8 & 11 & 12 & 1 & 13 \\ \dots & \dots & \dots & \dots & \dots \end{pmatrix}.$$

The equivalences are:

Augmented space element	$\Rightarrow$	$1\{\emptyset\}$	$2\{\emptyset\}$	$3\{\emptyset\}$	$4\{\emptyset\}$	$5\{\emptyset\}$	$1\{1\}$	$6\{\emptyset\}$
Discrete numbering	$\Rightarrow$	1	2	3	4	5	6	7
Augmented space element	$\Rightarrow$	$7\{\emptyset\}$	$8\{\emptyset\}$	$2\{2\}$	$9\{\emptyset\}$	$10\{\emptyset\}$	$3\{3\}$	
Discrete numbering	$\Rightarrow$	8	9	10	11	12	13.	

(B) *Reciprocal space formulation.* In reciprocal space the procedure is even simpler, since the operators act *only* on the configuration part of the space. As before, we start by numbering  $|k, \{\emptyset\}\rangle$  as 1 and then recursively generate the neighbours by acting on the states successively with  $\tilde{\mathbf{S}}$  and  $\tilde{\mathbf{F}}$ . Let us take the example of the square lattice:

- (i)  $\tilde{\mathbf{S}}$  acting on  $|\{\emptyset\}\rangle \equiv |1\rangle$  leaves it unchanged. The four neighbours of  $|1\rangle$  are then  $|1, |1\rangle, |1\rangle$  and  $|1\rangle$ .
- (ii)  $\tilde{\mathbf{F}}$  acting on  $|\{\emptyset\}\rangle \equiv |1\rangle$  gives  $|\{1\}\rangle$ . This we number  $|2\rangle$ .
- (iii)  $\tilde{\mathbf{S}}$  acting on  $|\{1\}\rangle \equiv |2\rangle$  gives  $|\{2\}\rangle, |\{3\}\rangle, |\{4\}\rangle$  and  $|\{5\}\rangle$ . These we number  $|3\rangle, |4\rangle, |5\rangle$  and  $|6\rangle$ .
- (iv)  $\tilde{\mathbf{F}}$  acting on  $|\{1\}\rangle \equiv |2\rangle$  gives  $|\{\emptyset\}\rangle \equiv |1\rangle$ .

We proceed as before and obtain the nearest neighbour map matrix. We again show below the initial part of the nearest neighbour map in reciprocal space:

$$\begin{pmatrix} 1 & 1 & 1 & 1 & 2 \\ 3 & 4 & 5 & 6 & 1 \\ 7 & 8 & 2 & 9 & 10 \\ \dots & \dots & \dots & \dots & \dots \end{pmatrix}.$$

Now the equivalences are:

Cardinality sequence	$\Rightarrow$	$\{\emptyset\}$	$\{1\}$	$\{2\}$	$\{3\}$	$\{4\}$	$\{5\}$	$\{6\}$	$\{7\}$	$\{8\}$	$\{1, 2\}$
Discrete numbering	$\Rightarrow$	1	2	3	4	5	6	7	8	9	10.

(C) *The block recursion.* We now go over to a matrix basis of the form  $\{\Phi_{J,LL'}^{(n)}\}$ , where  $J$  is the discrete labelling of the augmented space states and  $L, L'$  labels the angular momenta. The inner product of such a basis is defined by

$$(\Phi^{(n)}, \Phi^{(m)}) = \sum_J \sum_{L''} \Phi_{LL'',J}^{(n)\dagger} \Phi_{J,L''L'}^{(m)} = N_{LL'}^{nm}.$$

For a real space calculations on a lattice with  $Z$  nearest neighbours, we start the recursion with

$$\Phi_{J,LL'}^{(1)} = \mathbf{W}_{LL'}^{(1)} \delta_{J,1} + \mathbf{W}_{LL'}^{(2)} \delta_{J,Z+1},$$

while for a reciprocal space calculation we start with

$$\Phi_{J,LL'}^{(1)} = \mathbf{W}_{LL'}^{(1)} \delta_{J,1} + \mathbf{W}_{LL'}^{(2)} \delta_{J,2},$$

where

$$\mathbf{W}_{LL'}^{(1)} = \frac{A(\Delta_L^{-1/2})}{[A(\Delta_L^{-1})]^{1/2}} \delta_{LL'} \quad \mathbf{W}_{LL'}^{(2)} = \frac{F(\Delta_L^{-1/2})}{[A(\Delta_L^{-1})]^{1/2}} \delta_{LL'}. \quad (12)$$

The remaining terms in the basis are recursively obtained from

$$\begin{aligned} \sum_{L''} \Phi_{J,LL''}^{(2)} \mathbf{B}_{L''L'}^{(2)\dagger} &= \sum_{J'} \sum_{L''} H_{JL,J'L''} \Phi_{J',L''L'}^{(1)} - \sum_{L''} \Phi_{J,LL''}^{(1)} \mathbf{A}_{L''L'}^{(1)} \\ \sum_{L''} \Phi_{J,LL''}^{(n+1)} \mathbf{B}_{L''L'}^{(n+1)\dagger} &= \sum_{J'} \sum_{L''} H_{JL,J'L''} \Phi_{J',L''L'}^{(n)} - \sum_{L''} \Phi_{J,LL''}^{(n)} \mathbf{A}_{L''L'}^{(n)} - \sum_{L''} \Phi_{J,LL''}^{(n-1)} \mathbf{B}_{L''L'}^{(n)}. \end{aligned}$$

Orthogonalization of the basis gives

$$\sum_J \sum_{L''} \sum_{J'} \sum_{L'''} \Phi_{LL'',J}^{(n)\dagger} H_{JL',J'L''} \Phi_{J',L''L'''}^{(n)} = \sum_{L''} N_{LL''}^{nn} \mathbf{A}_{L''L'}^{(n)}. \quad (13)$$

In matrix notation, where the matrices are in angular momentum space,

$$\mathbf{A}^{(n)} = (\mathbf{N}^{nn})^{-1} \sum_J \sum_{J'} \Phi_J^{(n)\dagger} \mathbf{H}_{JJ'} \Phi_{J'}^{(n)}. \quad (14)$$

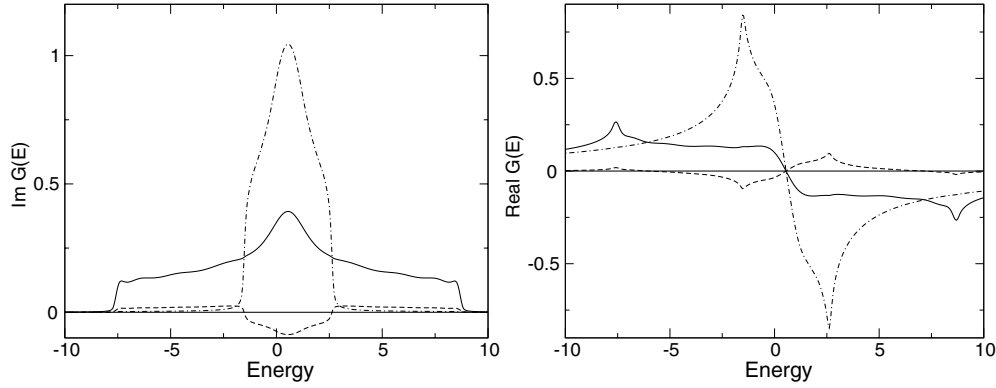
Next, we note that we started with  $J_{\max} \times L_{\max}^2$  orthogonal basis set. The above procedure merely gives  $J_{\max}$  basis sets. We still have orthogonality conditions among the various columns of  $\Phi_{J,LL'}^{(n)}$ . In order to impose these conditions, consider

$$\Psi_{J,LL'} = \sum_{J'} \sum_{L''} H_{JL,J'L''} \Phi_{J',L''L'}^{(n)} - \sum_{L''} \Phi_{J,LL''}^{(n)} \mathbf{A}_{L''L'}^{(n)} - \sum_{L''} \Phi_{J,LL''}^{(n-1)} \mathbf{B}_{L''L'}^{(n)}.$$

Construct  $L_{\max}$  column vectors  $\phi_{JL}^{(L)}$  out of the  $L_{\max}$  columns of  $\Psi_{J,LL'}$  and set about Gram–Schmidt orthonormalizing the set:

$$\begin{aligned} \phi_{LJ}^{(1)} &= \mathbf{B}_{11} \psi_{LJ}^{(1)} \Rightarrow \mathbf{B}_{11}^2 = \sum_{LJ} \phi_{JL}^{(1)} \phi_{LJ}^{(1)} \\ \phi_{LJ}^{(2)} &= \mathbf{B}_{21} \psi_{LJ}^{(1)} + \mathbf{B}_{22} \psi_{LJ}^{(2)} \Rightarrow \mathbf{B}_{21} = \sum_{LJ} \psi_{JL}^{(1)} \phi_{LJ}^{(2)}; \quad \mathbf{B}_{22}^2 = \sum_{LJ} \phi_{JL}^{(1)} \phi_{LJ}^{(1)} - \mathbf{B}_{21}^2 \\ &\dots \dots \dots \\ \phi_{LJ}^{(n)} &= \sum_{k=1}^n \mathbf{B}_{nk} \psi_{LJ}^{(k)} \Rightarrow \mathbf{B}_{nk} = \sum_{LJ} \psi_{JL}^{(k)} \phi_{LJ}^{(n)} \quad (k < n); \quad \mathbf{B}_{nn}^2 = \sum_{LJ} \phi_{JL}^{(n)} \phi_{LJ}^{(n)} - \sum_{k=1}^{n-1} \mathbf{B}_{nk}^2. \end{aligned} \quad (15)$$





**Figure 2.** The real and imaginary parts of the Green matrix for a  $2 \times 2$  Hamiltonian model. The full curves relate to  $\mathbf{G}_{11}$ , the dashed–dotted curve to  $\mathbf{G}_{22}$  and dashed curves to  $\mathbf{G}_{12}$ .

We may now construct  $\Phi_{J,LL'}^{(n+1)}$  from  $\psi_{JL}^{L'}$  and note that  $\mathbf{B}_{nk}$  is indeed the matrix  $\mathbf{B}^{(n+1)}$  that we are looking for.

Equations (14) and (15) show that we may calculate the matrices  $\{\mathbf{A}^{(n)}, \mathbf{B}^{(n+1)}\}$  recursively, noting that  $\mathbf{B}^{(1)} = \mathbf{I}$  and  $\mathbf{B}^{(0)} = \mathbf{0}$ . In this new basis, the Hamiltonian is *block tri-diagonal* and the Green matrix can be written as follows:

$$\mathbf{G}_{(n)} = [\mathbf{E}\mathbf{I} - \mathbf{A}^{(n)} - \mathbf{B}^{(n+1)\dagger} \mathbf{G}_{(n+1)} \mathbf{B}^{(n+1)}]^{-1} \quad (16)$$

$$\langle\langle \mathbf{G} \rangle\rangle = \mathbf{G}_{(1)}.$$

### 3. Model calculations

(A) *The model on a square lattice.* We shall first apply our methodology to a two-band model on a square lattice with the Hamiltonian

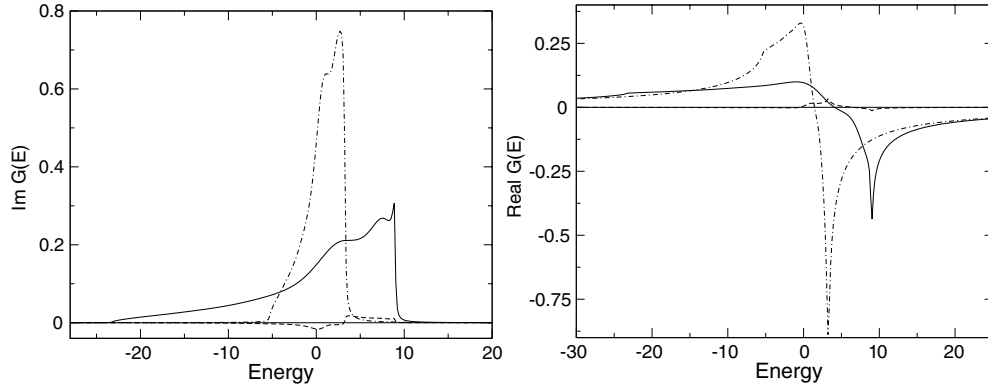
$$H^A = \sum_R \begin{pmatrix} 1.0 & 0.0 \\ 0.0 & 1.0 \end{pmatrix} P_R + \sum_R \sum_{R'} \begin{pmatrix} -2.0 & -0.2 \\ -0.2 & -0.5 \end{pmatrix} T_{RR'} \quad (17)$$

$$H^B = \sum_R \begin{pmatrix} 0.1 & 0.0 \\ 0.0 & 0.1 \end{pmatrix} P_R + \sum_R \sum_{R'} \begin{pmatrix} -2.0 & -0.2 \\ -0.2 & -0.5 \end{pmatrix} T_{RR'}.$$

The concentration is taken to be  $x = 0.5$ .

We have carried out block recursion for  $N = 10$  levels. The termination was carried out as suggested earlier by Godin and Haydock [7]: we put  $\{\mathbf{A}^{(n)}, \mathbf{B}^{(n+1)}\} = \{\mathbf{A}^{(N)}, \mathbf{B}^{(N+1)}\}$  for  $n = N + 1, \dots, N_{\max}$  and take  $\mathbf{G}_{(N_{\max}+1)} = (1/(E - i\delta))\mathbf{I}$ . In order to get a smooth density of states we had taken  $\delta = 0.01$  and  $N_{\max} = 10000$ . The elements of the Green matrix are shown in figure 2. The imaginary parts of the diagonal elements give the projected density of states. Herglotz properties of the diagonal parts give rise to a positive definite density of states. The off-diagonal part is relatively small and is not Herglotz. The projected density of states is symmetric, as is the imaginary part of the off-diagonal element. The real parts of the matrix elements are also shown. These are related to the imaginary parts by the Kramers–Krönig relation.

(B) *The s–d model of a transition metal–noble metal alloy.* Levin and Ehrenreich [22] and Gelatt and Ehrenreich [23] have introduced a simple two-band model for transition metal–noble metal alloys. Physical effects such as charge transfer between constituents will usually differ for the s–p conduction bands on one hand and the relatively narrow d bands on the other.



**Figure 3.** The real and imaginary parts of the Green matrix for the s-d model on a fcc lattice. The calculations were done by a real space block recursion. The full curves relate to  $\mathbf{G}_{11}$ , the dashed-dotted curve to  $\mathbf{G}_{22}$  and dashed curves to  $\mathbf{G}_{12}$ .

**Table 1.** Parameters for our calculation for an  $A_{50}B_{50}$  alloy on a fcc lattice.

Constituent	$\epsilon_s$	$\epsilon_d$	$\gamma$	$\alpha$	$t_d$
A	1.5	1.5	0.2	4.0	-0.5
B	0.0	0.0	0.2	4.0	-0.5

Their model includes the conduction bands described together as a single band and the set of d bands also described as a single band and their hybridization. The model also takes into account the large widths of the conduction bands and the relatively narrow widths of the d bands. The following Hamiltonian has many (but not all) of the essential features:

$$H = \sum_R \begin{pmatrix} \epsilon_s & \gamma \\ \gamma & \epsilon_d \end{pmatrix} P_R + \sum_R \sum_{R'} \begin{pmatrix} t_s & 0 \\ 0 & t_d \end{pmatrix} T_{RR'}. \quad (18)$$

The dominant disorder is taken to be in the terms  $\epsilon_s$  and  $\epsilon_d$ . The hybridization is taken between states at the same site only and the hopping terms are related by  $t_s = \alpha t_d$ . The sites  $R$  vary over the sites of a face-centred cubic lattice. As a model case we have taken the parameters shown in table 1.

(i) *Calculations using real space block recursion.* Figure 3 shows the real and imaginary parts of the Green matrix. These calculations were carried out through a real space block recursion technique. In general the qualitative features for the diagonal elements are similar to those of our square lattice model. The main difference is that on a fcc lattice the partial densities of states, related to the imaginary parts of the diagonal elements of the Green matrix, are no longer symmetric about the band centre. Consequently, the behaviour of the off-diagonal element is quite different. The total density of states is given by

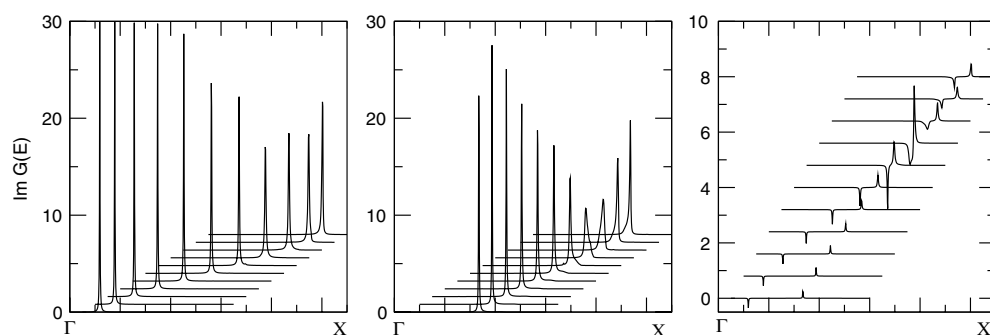
$$n(E) = (1/\pi) \text{Im}(G_{ss}(E - i\delta) + 5G_{dd}(E - i\delta)).$$

If, for example, the number of electrons in the constituents is 5 per atom per spin for A and 5.5 per atom per spin for B, the position of the Fermi energy is given by

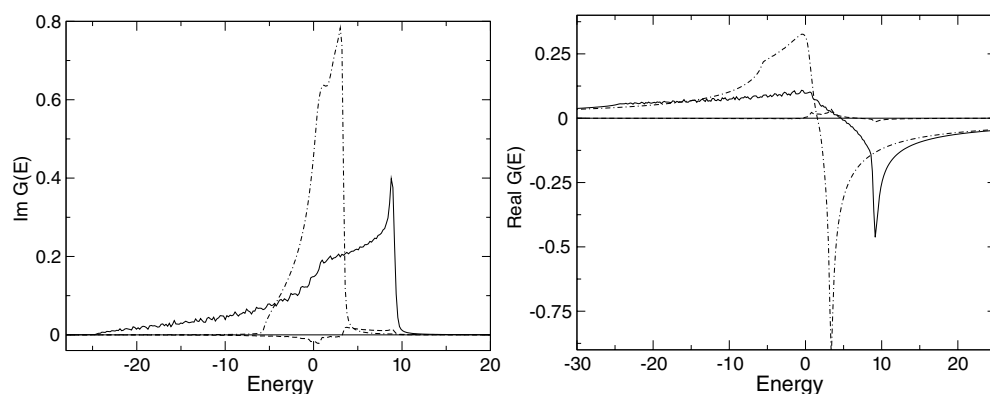
$$\int_{-\infty}^{E_F} dE n(E) = \langle n_e \rangle = x n_A + (1 - x) n_B = 5.25$$

where  $n_A$  and  $n_B$  are the numbers of valence electrons of the A and B types of atom.

(ii) *Calculations using reciprocal space block recursion.* We have carried out the block recursion in reciprocal space for the s-d model. The Green matrix in reciprocal space  $\mathbf{G}(\mathbf{k}, E)$



**Figure 4.** The matrix elements of  $\mathbf{G}(\mathbf{k}, E)$  on a fcc lattice: the 11 element (left), the 22 element (middle) and the 12 element (right) along the  $\Gamma$  to X direction.



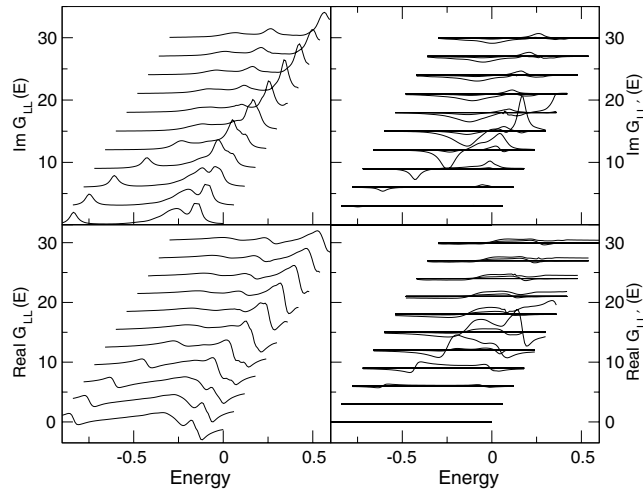
**Figure 5.** The real and imaginary parts of the Green matrix for the s-d model on a fcc lattice. The calculations are done by a  $k$ -space block recursion followed by a reciprocal space integration [25]. The full curves relate to  $\mathbf{G}_{11}$ , the dashed-dotted curve to  $\mathbf{G}_{22}$  and dashed curves to  $\mathbf{G}_{12}$ .

is the factor that arises in our earlier expression for the configuration averaged current-current correlation function. Its diagonal matrix element is related to the spectral functions for the different bands. In figure 4 we show the spectral functions along a given direction  $\Gamma$ -X in reciprocal space. The imaginary part of the off-diagonal matrix element is also shown in the figure. We note that the off-diagonal part of the Green matrix has antisymmetric structure in its peaks, while the diagonal matrix elements are positive (as they represent spectral functions).

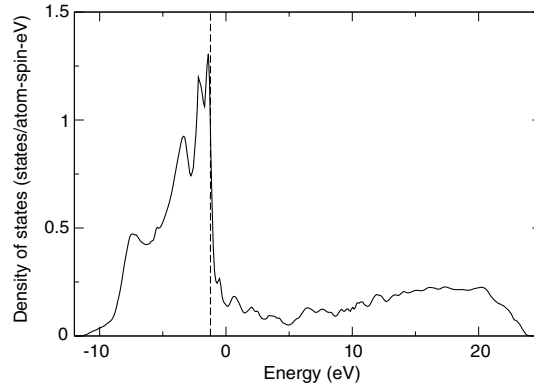
Figure 5 shows the Green matrix elements calculated starting from the  $\mathbf{G}(\mathbf{k}, E)$  and carrying out a reciprocal space integration developed by us [25] as a generalization of the tetrahedron method proposed by Jepsen and Andersen [24] for crystalline systems. The close comparison between figures 3 and 5 gives strong support for the accuracy of the reciprocal space recursion.

#### 4. Optical conductivity of $\text{Ni}_{50}\text{Pt}_{50}$ alloy

We shall now report a calculation of the optical conductivity of a 50–50 NiPt alloy. The initial electronic structure calculations require input of the potential parameters for pure Ni and Pt. These we have taken from a TB-LMTO calculation on the pure materials. We use these as input for a self-consistent calculation of the electronic structure using our LDA self-



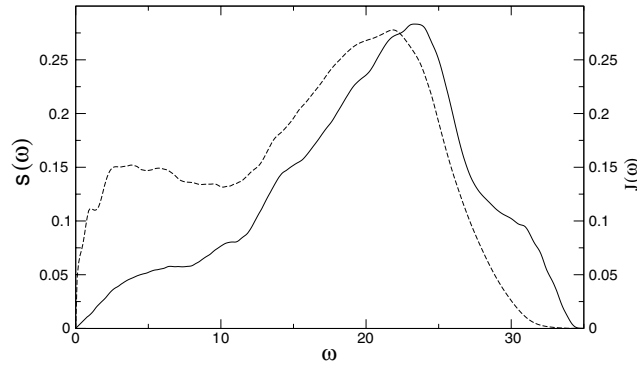
**Figure 6.** The imaginary and real parts of  $(1/9) \text{Tr } G_{LL}(\mathbf{k}, E)$  (left column) and the imaginary and real parts of  $G_{LL}'(E)$  (right column) for  $\text{Ni}_{50}\text{Pt}_{50}$  alloy.



**Figure 7.** The density of states for a 50–50 NiPt alloy. The dashed line shows the Fermi energy.

consistent augmented space recursion technique (ASR). We first calculate the Green matrix in reciprocal space. The results are shown in figure 6. The imaginary parts of the diagonal elements yield the spectral functions. Figure 6 also shows the off-diagonal matrix elements. Most of these elements are very small as compared with the spectral functions and are not positive definite as expected. The density of states for the alloy is shown in figure 7. We obtain the density of states by integrating the spectral function over the Brillouin zone, using the tetrahedron method generalized for disordered alloys by us [25]. The density of states for NiPt is particularly difficult to reproduce accurately by real space recursion, because of the sharp structure straddling the Fermi energy. However, the calculation in reciprocal space followed by Brillouin zone integration reproduces the density of states in very good agreement with earlier work using KKR-CPA [26].

The calculation of the optical conductivity involves the calculation of the four current terms  $\mathbf{j}^{AA}$ ,  $\mathbf{j}^{AB}$ ,  $\mathbf{j}^{BA}$  and  $\mathbf{j}^{BB}$ . Ideally one should calculate these for two atoms of the type AA, AB, BA or BB embedded in the disordered alloy. However, as a first approximation we have



**Figure 8.** The joint density of states  $J(\omega)$  (dashed curve) and the correlation function  $S(\omega)$  (solid curve) for 50–50 NiPt alloys.

obtained these for the pure metals and the ordered alloy. Having obtained the current terms we use the scattering methodology described in [4] to obtain the correlation function.

In figure 8 we show the result for the correlation function (full curves) for a 50–50 NiPt alloy:

$$S(\omega) = \frac{1}{4} \int dE_1 [S(E_1^+, E_2^+) + S(E_1^-, E_2^-) - S(E_1^+, E_2^-) - S(E_1^-, E_2^+)].$$

where  $E_2 = E_1 + \omega$ ,  $E^\pm = E \pm i0^+$ . For comparison we have also shown (as dashed curves) the joint density of states for the same alloy:

$$J(\omega) = \int dE n_v(E) n_c(E + \omega).$$

The former calculation requires the full Green matrices as well as the full matrix self-energies (matrices in the angular momentum space). We have carried out the calculations of these matrices via our block recursion technique. The joint density of states is modulated by the current terms. The main structures of the joint density of states are retained in the correlation function, but the relative heights of the principal peaks are modified.

## 5. Conclusion

In this communication we have described a block recursion in augmented space suitable for calculations of the Green matrices. The recursion is set up both with real space augmented by the configuration space of the alloy and the reciprocal space augmented with the configuration space. For the latter case we have coupled it with a Brillouin zone integration scheme which is a generalization of the tetrahedron method developed earlier for crystalline systems. The Green matrices are essential for the calculation of the response functions and effective current terms which are related to the self-energy matrices. We propose to use these techniques in our future applications.

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