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On the augmented-space cluster coherent-potential approximation and its analytic properties

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Abstract. We discuss the analytic properties of the configuration-averaged Green function obtained using the self-consistent cluster coherent-potential approximation, developed in the augmented-space formalism. It is shown that the iteration scheme for the self-energy is always convergent to a unique value, which is bounded and *herglotz*, provided we start off with a bounded and *herglotz* guess. This ensures that we shall always get a unique, non-negative density of states at all energies.

1. Introduction

Our understanding of the electronic properties of random, substitutionally disordered alloys has increased greatly since the introduction and application of the coherent-potential approximation (CPA) (Soven 1967, Taylor 1967, Velicky *et al* 1968). The CPA yields a unique and analytic averaged Green function and guarantees a non-negative density of states (Muller-Hartman 1973, Ducastelle 1974). In spite of all the successes it has achieved, the CPA, being a single-site approximation, cannot be used to investigate local environmental effects, which involve correlations among many sites.

The idea of extending the CPA to include the effects of clusters, short-range order and off-diagonal disorder has been mooted by several authors in both the tight-binding and Korringa–Kohn–Rostoker (KKR) frameworks (Nickel and Krumhansl 1971, Tsukada 1969, 1972, Bishop and Mookerjee 1974, Kaplan *et al* 1980, Mills and Ratanavararaksa 1978, Mills *et al* 1983, Kaplan and Gray 1984, Mookerjee 1987a, Razee *et al* 1990). One of the major difficulties with most generalizations is the violation of the analytic properties of the approximated averaged Green function (Nickel and Butler 1973), thereby giving unphysical results. The successful generalizations of the CPA that have been proved to be analytic are the travelling cluster approximation (Mills and Ratanavararaksa 1978, Kaplan *et al* 1980) and the molecular CPA (Tsukada 1972). A simpler generalization of the CPA is the cluster coherent-potential approximation (CCPA) (Kumar *et al* 1982, Mookerjee 1987b), which is developed in the augmented-space formalism (ASF) (Mookerjee 1973, Kaplan and Gray 1976a, b). The exact nature of this approximation and its analytic properties need explicit discussion. This is to put to rest doubts about the analytic properties of the CCPA as well as loose statements about the

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travelling cluster approximation being the *only* possible generalization of the single-site CPA.

The outline of this paper is as follows. In section 2 we present the basic formulation. In section 2.1 we outline the analytic properties of the Green function and the self-energy in the exact case. In section 2.2 we briefly discuss the ASF and in section 2.3 we derive the CCPA equations. In section 3 we prove three propositions regarding the analyticity of the CCPA; we show that the density of states is always positive within the CCPA and that the iteration scheme for the self-energy always converges to a unique and bounded value if we start the iteration with a bounded guess. Finally, a conclusion is given in section 4.

2. Formulation of the problem

There is considerable confusion over the notations used in the discussions of the augmented space. A description of configuration fluctuations of very large systems is intrinsically difficult. To make it easily comprehensible, we shall follow a hybrid between the notations of Kaplan *et al* (1980) and our own earlier notations.

2.1. General properties of the Green functions

We consider a random tight-binding Hamiltonian $\mathbf{H}$, given by

$$\mathbf{H} = \sum_i \varepsilon_i |i\rangle\langle i| + \sum_{\substack{i,j \\ (i\neq j)}} V_{ij} |i\rangle\langle j|. \tag{2.1}$$

The states $\{|i\rangle\}$ are Wannier states associated with the sites $\{i\}$. The Green function of this system is given by

$$\mathbf{G}(z) = (z\mathbf{I} - \mathbf{H})^{-1} \tag{2.2}$$

where z is a complex number. In this paper, we will always work on the upper half-plane, $\operatorname{Im} z > 0$. The imaginary part of an operator $\mathbf{A}$ is defined by

$$\operatorname{Im} \mathbf{A} = (\mathbf{A} - \mathbf{A}^\dagger)/2\mathrm{i} \tag{2.3}$$

where $\mathbf{A}^\dagger$ is the Hermitian adjoint of $\mathbf{A}$. It then follows from equation (2.2) that

$$\operatorname{Im} \mathbf{G}(z) = -\mathbf{G}^\dagger(z)\mathbf{G}(z) \operatorname{Im} z \tag{2.4}$$

where

$$\mathbf{G}^\dagger(z) = \mathbf{G}(z^*).$$

Since $\mathbf{H}$ is real and bounded, it follows from equations (2.2) and (2.4) that $\mathbf{G}(z)$ is a *herglotz* analytic function, i.e.

(i) $\mathbf{G}(z)$ is analytic everywhere except for parts of the real z axis,
(ii) its spectrum is bounded and $\mathbf{G}(z)$ behaves like $1/z$ as $z \to \pm\infty$, and
(iii) $\operatorname{Im} \mathbf{G}(z) < 0$ for $\operatorname{Im} z > 0$.

The configuration-averaged Green function is given by

$$\langle \mathbf{G}(z) \rangle \equiv \bar{\mathbf{G}}(z) = (z\mathbf{I} - \mathbf{\Sigma})^{-1}. \tag{2.5}$$

The operator $\mathbf{\Sigma}$ is called the self-energy operator. Averaging equation (2.4) we get

$$\operatorname{Im} \bar{\mathbf{G}}(z) = -\langle \mathbf{G}(z)\mathbf{G}^{\dagger}(z) \rangle \operatorname{Im} z. \tag{2.6}$$

Therefore, the averaged Green function $\bar{\mathbf{G}}(z)$ retains the *herglotz* analytic properties of the exact Green function $\mathbf{G}(z)$. Again, from equation (2.5) we get

$$\operatorname{Im} \bar{\mathbf{G}}(z) = -\bar{\mathbf{G}}(z) \operatorname{Im}(z\mathbf{I} - \mathbf{\Sigma})\bar{\mathbf{G}}^{\dagger}(z) \tag{2.7}$$

which implies that $\operatorname{Im} \mathbf{\Sigma}$ is negative definite (Ducastelle 1974). For any approximation to give physically acceptable results, the approximated averaged Green function and the self-energy must satisfy the *herglotz* properties.

2.2. *The augmented-space formalism*

Let $F(\{n_i\})$ be a function of a set of random variables $\{n_i\}$, whose probability densities $p_i(n_i)$ have finite moments to all orders. Thus $p_i(n_i)$ can always be written as a continued-fraction expansion (Shohat and Tamarkin 1963), which means that we can write $p_i(n_i)$ as the matrix element

$$p_i(n_i) = -(1/\pi) \operatorname{Im}\langle \nu_0^i | [(n_i + \mathrm{i}0)\mathbf{I} - \mathbf{M}_i]^{-1} | \nu_0^i \rangle$$

where $\mathbf{M}_i$ is an operator in the vector space ϕ_i, called the configuration space. The representation of $\mathbf{M}_i$ in ϕ_i is a tridiagonal matrix. The ground configuration state $|\nu_0^i\rangle$ is such that $\langle \nu_0^i | \mathbf{M}_i | \nu_0^i \rangle$ is the first element in the continued-fraction expansion (Mookerjee 1973). Note that the vector space ϕ_i is spanned by the different configuration states $\{|\nu_m^i\rangle\}$, where m is the cardinality of the basis (Paquet and Leroux-Hugon 1984).

The augmented-space theorem (Mookerjee 1973) then tells us that the configuration average of $F(\{n_i\})$ is given by

$$\langle F(\{n_i\}) \rangle = \langle f | \tilde{F}(\{\mathbf{M}_i\}) | f \rangle \tag{2.8}$$

where $\tilde{F}(\{\mathbf{M}_i\})$ is an operator functional in the augmented space $\Psi = \mathcal{H} \otimes \Phi$. Here $\mathcal{H}$ is the real Hilbert space spanned by $\{|i\rangle\}$ and $\Phi = \Pi_i \phi_i$ is the total configuration space. The state $|f\rangle = \Pi_i |\nu_0^i\rangle$ is the ground configuration state in the total configuration space Φ. A general configuration state spanning Φ is of the form

$$|f_{\sigma,C}\rangle = |\nu_{m_1}^1 \, \nu_{m_2}^2 \, \nu_{m_3}^3 \ldots \nu_{m_i}^i \ldots \rangle$$

and can be uniquely described by the set of points $\sigma = \{j: m_j \neq 0\}$ at which there are excitations and the set of cardinalities $C = \{m_j\}$ at those points (Paquet and Leroux-Hugon 1984). In the case of binomial distributions (as for binary alloys) the cardinality of any excitation is 1 and this labelling C may be omitted. In the following, unless essential, we shall omit the cardinality label. In this notation, the ground configuration state $|f\rangle \equiv |f_{\varnothing}\rangle$, where $\varnothing$ is the null set. We shall also write $\langle f_{\sigma} | \tilde{\mathbf{\Omega}} | f_{\sigma'} \rangle$ as $\mathbf{\Omega}^{\sigma\sigma'}$. In the above notation, the augmented-space theorem may be written as

$$\langle F(\{n_i\}) \rangle = [F(\{\mathbf{M}_i\})]^{\varnothing\varnothing}. \tag{2.9}$$

The disordered Hamiltonian of equation (2.1) is a function of the random variables,

i.e. $\mathbf{H} \equiv \mathbf{H}(\{n_i\})$. By the augmented-space theorem, the configuration-averaged Green function can be written as

$$\langle \mathbf{G}(z) \rangle = [\![[z\tilde{\mathbf{I}} - \tilde{\mathbf{H}}(\{\mathbf{M}_i\})]^{-1}]\!]^{\varnothing\varnothing} = [\tilde{\mathbf{G}}(\{\mathbf{M}_i\})]^{\varnothing\varnothing} \tag{2.10}$$

where $\tilde{\mathbf{H}}(\{\mathbf{M}_i\})$ is the augmented Hamiltonian acting on the augmented space and constructed by replacing the n_i in $\mathbf{H}(\{n_i\})$ by the corresponding $\mathbf{M}_i$ (Mookerjee 1973). Note that our discussion includes off-diagonal disorder (Kaplan and Gray 1976a) and Markovian short-range order (Gray and Kaplan 1981) in the construction of the operator $\mathbf{M}_i$. In the case where disorder is homogeneous, i.e. if $p_i(n_i)$ is independent of the label i, the augmented Hamiltonian has the following translational symmetry in the augmented space Ψ:

$$\text{if } \mathcal{T} \equiv [i \to i+n;\ \sigma \equiv (i,j,\ldots) \to \sigma + n \equiv (i+n, j+n, \ldots);\ |f_\sigma\rangle \to |f_{\sigma+n}\rangle]$$

$$\text{then} \qquad \mathcal{T}\tilde{\mathbf{H}} = \tilde{\mathbf{H}}.$$

In any approximation, the self-energy should also satisfy this translational symmetry.

2.3. *The cluster coherent-potential approximation (CCPA)*

Equation (2.10) is an exact expression, and therefore will yield a *herglotz* averaged Green function. But we observe that the rank of the augmented space is $N \times 2^N$, and hence it is not possible to use equation (2.10) for computational purposes, since N is very large. Thus the CCPA is used to overcome this difficulty. In the following we analyse the procedure leading to the CCPA.

We choose a finite compact cluster $\mathbb{C}$ of sites in $\mathcal{H}$. It is immaterial which particular set of sites we choose. What is important is the relative position of these sites with respect to one another. Let $\mathscr{C}$ be the set of configurations of this cluster.

In the first step, we define a subspace Ψ_1 spanned by $\{|i, f_\sigma\rangle,\ i \in \mathbb{C},\ f_\sigma \in \mathscr{C}\}$ and its complement in Ψ, $\Psi_2 = \Psi \backslash \Psi_1$. If we partition Ψ into these two subspaces, the partition theorem gives

$$\mathbf{G}_1 = (z\mathbf{I}_1 - \mathbf{H}_1 - \mathbf{H}_{12}\mathbf{G}_2\mathbf{H}_{12}^\dagger)^{-P_1} = (z\mathbf{I}_1 - \hat{\mathbf{H}})^{-P_1} \tag{2.11}$$

where

$$\hat{\mathbf{H}} = \mathbf{H}_1 + \mathbf{H}_{12}\mathbf{G}_2\mathbf{H}_{12}^\dagger \qquad \mathbf{H}_1 = \mathcal{P}_1\tilde{\mathbf{H}}\mathcal{P}_1 \qquad \mathbf{H}_2 = \mathcal{P}_2\tilde{\mathbf{H}}\mathcal{P}_2$$

$$\mathbf{H}_{12} = \mathcal{P}_1\tilde{\mathbf{H}}\mathcal{P}_2 \qquad \mathbf{G}_2 = (z\mathbf{I}_2 - \mathbf{H}_2)^{-P_2}$$

$\mathcal{P}_j$ is the projection operator in the subspace labelled by j and $\mathbf{X}^{-P_j}$ is the inverse of the operator $\mathbf{X}$ in the subspace labelled by *j alone*. If we write $\hat{\mathbf{H}} = \mathbf{H}_1 + \boldsymbol{\xi}$, then we have

$$\xi^{\sigma\sigma'} = \sum_{\alpha,\alpha'} \mathbf{H}_{12}^{\sigma\alpha}\mathbf{G}_2^{\alpha\alpha'}\mathbf{H}_{12}^{\dagger\alpha'\sigma'}. \tag{2.12}$$

We shall now introduce the crucial approximation on $\boldsymbol{\xi}$, in which the subspace Ψ_2 is replaced by an effective medium and therefore has only one configuration, namely the ground state $|f\rangle$. Note that the effective medium is translationally symmetric. It is defined by the self-energy $\boldsymbol{\Sigma}$, which is the effective Hamiltonian. Therefore, we have

$$\mathbf{G}_2^{\alpha\alpha'} = \mathbf{G}_2(\boldsymbol{\Sigma})\delta_{\alpha\alpha'}\delta_{\alpha\varnothing} \qquad \mathbf{H}_{12}^{\sigma\alpha} = \boldsymbol{\Sigma}_{12}\delta_{\alpha\varnothing} \qquad \mathbf{G}_2(\boldsymbol{\Sigma}) = (z\mathbf{I}_2 - \boldsymbol{\Sigma}_2)^{-P_2} \tag{2.13}$$

where $\boldsymbol{\Sigma}_2$ and $\boldsymbol{\Sigma}_{12}$ are $m \times m$ and $n \times m$ matrices made by partitioning the self-energy

Σ, such that the cluster has n sites and $m = N - n$, N being the system size. Therefore, equation (2.12) now reads

$$\xi^{\sigma\sigma'} = \Sigma_{12}\mathbf{G}_2\Sigma_{12}^{\dagger}\delta_{\sigma\sigma'}. \tag{2.14}$$

We note that $\mathbf{G}_2 = \bar{\mathbf{G}}^{(\mathbb{C})}$, i.e. it is the Green function of the effective medium, calculated on the lattice from which the cluster $\mathbb{C}$ has been removed.

In the second step, we partition Ψ_1 into a subspace ψ_1, spanned by $\{|i,f\rangle, i \in \mathbb{C}\}$, and its complement ψ_2, spanned by $\{|i,f_\sigma\rangle, \sigma \neq \varnothing, i \in \mathbb{C}\}$. The partition theorem and the augmented-space theorem together yield

$$\bar{\mathbf{G}} = \mathbf{G}_{\psi_1} = (z\mathbf{I} - \mathbf{h}_1 - \mathbf{h}_{12}\Gamma\mathbf{h}_{12}^{\dagger})^{-P_{\psi_1}} \tag{2.15}$$

where

$$\mathbf{h}_1 = \mathcal{P}_{\psi_1}\hat{\mathbf{H}}\mathcal{P}_{\psi_1} \qquad \mathbf{h}_{12} = \mathcal{P}_{\psi_1}\hat{\mathbf{H}}\mathcal{P}_{\psi_2}$$
$$\Gamma = (z\mathbf{I} - \mathbf{h}_2)^{-P_{\psi_2}} \qquad \mathbf{h}_2 = \mathcal{P}_{\psi_2}\hat{\mathbf{H}}\mathcal{P}_{\psi_2}.$$

Here $\mathcal{P}_{\psi_i}$ is the projection operator in the subspace ψ_i and is given by

$$\mathcal{P}_{\psi_1} = \sum_{i\in\mathbb{C}} |i,f\rangle\langle i,f| \qquad \mathcal{P}_{\psi_2} = \sum_{i\in\mathbb{C}}\sum_{\sigma\neq\varnothing} |i,f_\sigma\rangle\langle i,f_\sigma|.$$

Note that

$$\mathbf{h}_{12}^{\varnothing\sigma'} = \hat{\mathbf{H}}^{\varnothing\sigma'}\delta_{\sigma\varnothing} = \mathbf{H}_1^{\varnothing\sigma'}$$

which is independent of Σ, since subspace Ψ_1 contains the real cluster only. Thus we write

$$\mathbf{h}_{12}^{\varnothing\sigma'} = \mathbf{K} \qquad \mathbf{h}_1 = \hat{\mathbf{H}}^{\varnothing\varnothing} = \mathbf{H}_1^{\varnothing\varnothing} + \xi^{\varnothing\varnothing}. \tag{2.16}$$

It should be noted that $\mathbf{H}_1^{\varnothing\varnothing} = \mathbf{H}_{\mathrm{vc}}$, the virtual crystal Hamiltonian.

Now, for a translationally symmetric effective medium, the Green function in the subspace ψ_1 can be written as

$$\mathbf{G}_{\psi_1}^{\mathrm{eff}} = (z\mathbf{I} - \Sigma)^{-P_{\psi_1}} = (z\mathbf{I} - \Sigma_{\psi_1} - \xi^{\varnothing\varnothing})^{-1}. \tag{2.17}$$

Here, Σ is the self-energy of the full effective medium, and thus is an N-dimensional matrix, while Σ_{ψ_1} is n-dimensional, where n is the size of the cluster.

The CCPA equations are obtained by equating equations (2.15) and (2.17) as

$$\Sigma = \mathbf{H}_{\mathrm{vc}} + \mathbf{K}\Gamma(\Sigma)\mathbf{K}^{\dagger} = \mathcal{F}(\Sigma). \tag{2.18}$$

In equation (2.18), we have suppressed the subscript ψ_1 in Σ. This subscript is not essential since equation (2.18) is solved only in the cluster subspace.

3. Analytic properties of the CCPA Green functions

Let us now restrict z to a compact subspace $\mathcal{C}$ of the upper half-plane such that, whenever $z \in \mathcal{C}$, $\|\mathbf{H}_{\mathrm{vc}}\| \leqslant X$ and $\|\mathbf{K}\| \leqslant Y$.

Proposition 1. If $\operatorname{Im}\Sigma < 0$ in $\mathcal{C}$, then $\operatorname{Im}\mathcal{F}(\Sigma) < 0$.

Proof. We shall first show that, if $\mathbf{A} = \mathbf{BCB}^\dagger$, then Im $\mathbf{A}$ and Im $\mathbf{C}$ have the same sign. We have

$$\text{Im}\,\mathbf{A} = (\mathbf{A} - \mathbf{A}^\dagger)/2\mathrm{i} = (1/2\mathrm{i})\mathbf{B}(\mathbf{C} - \mathbf{C}^\dagger)\mathbf{B}^\dagger = \mathbf{B}(\text{Im}\,\mathbf{C})\mathbf{B}^\dagger$$

from which the result follows.

Let us call $\mathbf{G}_2^{\varnothing\varnothing} = \boldsymbol{\gamma}$ and $\xi^{\sigma\sigma} = \xi$. Then $\boldsymbol{\gamma}^{-1} = z\mathbf{I} - \boldsymbol{\Sigma}$, so that Im $\boldsymbol{\gamma} < 0$ if Im $\boldsymbol{\Sigma} < 0$ (since Im $z > 0$). Again, from equation (2.14) we get

$$\text{Im}\,\xi = \boldsymbol{\Sigma}_{12}(\text{Im}\,\boldsymbol{\gamma})\boldsymbol{\Sigma}_{12}^\dagger < 0. \tag{3.1}$$

Therefore, $\hat{\mathbf{H}} = \mathbf{H}_1 + \xi \Rightarrow \text{Im}\,\hat{\mathbf{H}} < 0$, since $\mathbf{H}_1$ is real. Again, we have

$$\mathbf{h}_2^{\sigma\sigma'} = \mathbf{H}_1^{\sigma\sigma'} + \xi^{\sigma\sigma'}\delta_{\sigma\sigma'} \qquad \boldsymbol{\Gamma}^{-1} = z\mathbf{I} - \mathbf{h}_2 \tag{3.2}$$

which imply

$$\text{Im}\,\mathbf{h}_2 = \text{Im}\,\xi < 0 \qquad \text{Im}\,\boldsymbol{\Gamma} < 0.$$

From equation (2.18) we get the required result

$$\text{Im}\,\mathfrak{F}(\boldsymbol{\Sigma}) = \text{Im}[\mathbf{K}\boldsymbol{\Gamma}(\boldsymbol{\Sigma})\mathbf{K}^\dagger] = \mathbf{K}[\text{Im}\,\boldsymbol{\Gamma}(\boldsymbol{\Sigma})]\mathbf{K}^\dagger < 0.$$

Now let us consider the CCPA Green function as given in equation (2.15), which gives

$$\bar{\mathbf{G}}^{-1} = z\mathbf{I} - \mathbf{h}_1 - \mathbf{K}\boldsymbol{\Gamma}\mathbf{K}^\dagger.$$

We have seen that for Im $\boldsymbol{\Sigma} < 0$, Im $\mathbf{h}_1 < 0$ and Im $\boldsymbol{\Gamma} < 0$, which readily proves that

$$\text{Im}\,\bar{\mathbf{G}} < 0 \qquad \text{in the plane Im}\,z > 0$$

and therefore guarantees positive density of states.

To understand this, we illustrate it with the CCPA for a two-atom cluster (Mookerjee 1987b). We take the cluster $\mathbb{C} \equiv \{0, 1\}$ and hence $\mathscr{C} \equiv \{f, f_0, f_1, f_{01}\}$. For no off-diagonal disorder, the matrices $\mathbf{h}_1$ and $\mathbf{h}_{12}$ are given by

$$\mathbf{h}_1 = \begin{pmatrix} \bar{\varepsilon} + \xi_{00} & V_{01} + \xi_{01} \\ V_{10} + \xi_{10} & \bar{\varepsilon} + \xi_{00} \end{pmatrix}$$

$$\mathbf{h}_{12} = \begin{pmatrix} \omega & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & \omega \end{pmatrix}$$

where

$$\bar{\varepsilon} = x\varepsilon_A + y\varepsilon_B \qquad \omega = (xy)^{1/2}(\varepsilon_A - \varepsilon_B)$$

and x and y are the respective concentrations of the constituents A and B. The matrix $\mathbf{h}_2$ is a 6×6 tridiagonal matrix and thus $\boldsymbol{\Gamma}$ can be obtained easily. The averaged Green function can then be written as (Rajput *et al* 1990)

$$\langle G_{00}\rangle = [z - \bar{\varepsilon} - \xi_{00} - \omega R_6^{-1}\omega - (V_{01} + \xi_{01} + \omega Q\omega)(z - \bar{\varepsilon} - \xi_{00} - \omega R_6^{-1}\omega)^{-1} \times (V_{10} + \xi_{10} + \omega Q^\dagger\omega)]^{-1} \tag{3.3}$$

where

$$\bar{\varepsilon} = x\varepsilon_B + y\varepsilon_A$$

$$R_6 = z - \bar{\varepsilon} - \xi_{00} - (V_{10} + \xi_{10})R_5^{-1}(V_{01} + \xi_{01}) \qquad R_5 = z - \bar{\varepsilon} - \xi_{00} - \omega R_4^{-1}\omega$$

$$R_4 = z - \tilde{\varepsilon} - \xi_{00} - (V_{01} + \xi_{01})R_3^{-1}(V_{10} + \xi_{10}) \qquad R_3 = z - \tilde{\varepsilon} - \xi_{00} - \omega R_2^{-1}\omega$$

$$R_2 = z - \tilde{\varepsilon} - \xi_{00} - (V_{10} + \xi_{10})R_1^{-1}(V_{01} + \xi_{01}) \qquad R_1 = z - \tilde{\varepsilon} - \xi_{00}$$

and

$$Q = R_6^{-1}(V_{01} + \xi_{01})R_5^{-1}\omega R_4^{-1}(V_{10} + \xi_{10})R_3^{-1}\omega R_2^{-1}(V_{01} + \xi_{01})R_1^{-1}.$$

We note that $\operatorname{Im} R_1 = -\operatorname{Im} \xi_{00} > 0$ and this property is common to all the R_j, i.e. $\operatorname{Im} R_j > 0, j = 1, 2, \ldots, 6$. Therefore, from equation (3.3) we get

$$\operatorname{Im} \langle G_{00}(z) \rangle < 0.$$

Proposition 2. If $\|\boldsymbol{\Sigma}\|$ is bounded, so is $\|\mathcal{F}(\boldsymbol{\Sigma})\|$.

Proof. We shall use the inequality that, if $\operatorname{Im} \mathbf{A} \geqslant x > 0$, then $\|\mathbf{A}^{-1}\| \leqslant 1/x$ (Kaplan *et al* 1980). We know that $\operatorname{Im}(z\mathbf{I} - \boldsymbol{\Sigma}) \geqslant x > 0$ for $z \in \mathcal{C}$, and for $\operatorname{Im} \boldsymbol{\Sigma} < 0$, this implies that $\|\boldsymbol{\gamma}\| \leqslant 1/x$. Hence, from equation (2.14) we get $\|\boldsymbol{\xi}\| < Y$, where $Y = q^2/x$, and $\|\boldsymbol{\Sigma}\| < q$. Since $\mathbf{H}_{vc}$ is bounded above by X, we have $\|\hat{\mathbf{H}}\| \leqslant X + Y$, also bounded above.

Now from equation (2.18) we note that

$$\|\mathcal{F}(\boldsymbol{\Sigma})\| \leqslant \|\mathbf{H}_{vc}\| + \|\mathbf{K}\|^2 \|\boldsymbol{\Gamma}(\boldsymbol{\Sigma})\|.$$

But since $\operatorname{Im} \boldsymbol{\Gamma}^{-1} \geqslant r > 0$ (see proof of proposition 1), $\|\boldsymbol{\Gamma}(\boldsymbol{\Sigma})\| \leqslant 1/r$, so that we have

$$\|\mathcal{F}(\boldsymbol{\Sigma})\| < X + Y^2/r$$

and thus is bounded.

Proposition 3. The iterative chain $\boldsymbol{\Sigma}^{n+1} = \mathcal{F}(\boldsymbol{\Sigma}^n)$ converges to a unique solution, provided $-\operatorname{Im} \boldsymbol{\xi}^n \geqslant X > 0$ and $-\operatorname{Im} \boldsymbol{\Sigma}^n \geqslant Y > 0$ at each stage of the iteration and $\boldsymbol{\Sigma}^0$, $\mathbf{K}$ and $\mathbf{G}^0$ are bounded to start with.

Proof. The iteration scheme for the self-energy can be written from equation (2.18) as

$$\boldsymbol{\Sigma}^m = \mathcal{F}(\boldsymbol{\Sigma}^{m-1}) = \mathbf{H}_{vc} + \mathbf{K}\boldsymbol{\Gamma}^{m-1}\mathbf{K}^\dagger \tag{3.4}$$

where

$$\boldsymbol{\Gamma}^m = (z\mathbf{I} - \mathbf{H}_1 - \boldsymbol{\xi}^m)^{-1}$$

and m is the number of iterations. We can rewrite equation (2.14) as

$$\boldsymbol{\xi} = \boldsymbol{\Sigma}\mathbf{G}\boldsymbol{\Sigma}^\dagger.$$

Here we have suppressed the subspace indices in $\boldsymbol{\Sigma}$ and $\mathbf{G}$ for convenience, and these may be restored wherever appropriate.

For the proof, we shall pick only the even steps in the iteration, i.e. for m even,

$$\boldsymbol{\xi}^{m-1} = \boldsymbol{\Sigma}^{m-2}\mathbf{G}^{m-2}(\boldsymbol{\Sigma}^{m-3})^\dagger \qquad \boldsymbol{\xi}^m = \boldsymbol{\Sigma}^{m-2}\mathbf{G}^{m-1}(\boldsymbol{\Sigma}^{m-3})^\dagger \tag{3.5}$$

where

$$\mathbf{G}^m = (z\mathbf{I} - \boldsymbol{\Sigma}^m)^{-1}.$$

Thus our iteration scheme is as follows:

$$\boldsymbol{\Sigma}^0 \to \boldsymbol{\xi}^1 = \boldsymbol{\Sigma}^0\mathbf{G}^0(\boldsymbol{\Sigma}^{-1})^\dagger \to \boldsymbol{\Sigma}^1 \to \boldsymbol{\xi}^2 = \boldsymbol{\Sigma}^0\mathbf{G}^1(\boldsymbol{\Sigma}^{-1})^\dagger \to \boldsymbol{\Sigma}^2 \to \boldsymbol{\xi}^3$$

$$= \boldsymbol{\Sigma}^2\mathbf{G}^2(\boldsymbol{\Sigma}^1)^\dagger \to \boldsymbol{\Sigma}^3 \to \boldsymbol{\xi}^4 = \boldsymbol{\Sigma}^2\mathbf{G}^3(\boldsymbol{\Sigma}^1)^\dagger \to \text{and so on}$$

where $\boldsymbol{\Sigma}^0 = \boldsymbol{\Sigma}^{-1}$ is our starting self-energy, which we assume to be bounded and *herglotz*.

From equations (3.4) and (3.5) we have

$$\Delta\xi^m = \xi^m - \xi^{m-1} = \Sigma^{m-2}\mathbf{G}^{m-1}\Delta\Sigma^{m-1}\mathbf{G}^{m-2}(\Sigma^{m-3})^\dagger \tag{3.6a}$$

and

$$\Delta\Sigma^m = \Sigma^m - \Sigma^{m-1} = \mathbf{K}\Gamma^{m-1}\Delta\xi^{m-1}\Gamma^{m-2}\mathbf{K}^\dagger. \tag{3.6b}$$

Since $\mathrm{Im}(\mathbf{G}^m)^{-1} \geqslant Y' > 0$, we can define $\mathrm{Im}(\mathbf{G}^m)^{-1} = (1/\alpha^m)^2$ and hence equation (3.6*a*) can be written as

$$\Delta\xi^m = (\mathbf{Y}^{m-1})^\dagger\alpha^{m-1}\Delta\Sigma^{m-1}\alpha^{m-2}\mathbf{Y}^{m-2} \tag{3.7}$$

with

$$\mathbf{Y}^m = \mathbf{G}^m(\Sigma^{m-1})^\dagger/\alpha^m.$$

We shall now polar decompose $\mathbf{Y}^m$ as $\mathbf{W}^m\mathbf{J}^m$ (Halmos 1967) where $\mathbf{J}^m = [(\mathbf{Y}^m)^\dagger\mathbf{Y}^m]^{1/2}$ and $\mathbf{W}^m$ is a partial isometry from the range of $\mathbf{J}^m$ to the range of $\mathbf{Y}^m$ and thus $\|\mathbf{W}^m\|^2 = 1$. With this we can write equation (3.7) as

$$\Delta\xi^m = \mathbf{J}^{m-1}[(\mathbf{W}^{m-1})^\dagger\alpha^{m-1}\Delta\Sigma^{m-1}\alpha^{m-2}\mathbf{W}^{m-2}]\mathbf{J}^{m-2}. \tag{3.8}$$

In a similar manner, defining $\mathrm{Im}(\Gamma^m)^{-1} = (1/\beta^m)^2 \geqslant X' > 0$, we may write equation (3.6*b*) as

$$\Delta\Sigma^m = \mathcal{J}^{m-1}[(\mathbf{W}^{m-1})^\dagger\beta^{m-1}\Delta\xi^{m-1}\beta^{m-2}\mathbf{W}^{m-2}]\mathcal{J}^{m-2} = \mathcal{J}^{m-1}\mathcal{A}^{m-1}\mathcal{J}^{m-2} \tag{3.9}$$

where

$$\begin{aligned} &\mathcal{J}^m = [(\mathcal{Y}^m)^\dagger\mathcal{Y}^m]^{1/2} \qquad \text{with} \qquad \mathcal{Y}^m = \Gamma^m\mathbf{K}^\dagger/\beta^m \\ &\mathcal{A}^m = (\mathbf{W}^m)^\dagger\beta^m\Delta\xi^m\beta^{m-1}\mathbf{W}^{m-1}. \end{aligned} \tag{3.10}$$

With the help of equations (3.8) and (3.9) we can write equation (3.10) as

$$\mathcal{A}^m = [(\mathbf{W}^m)^\dagger\beta^m\mathbf{J}^{m-1}(\mathbf{W}^{m-1})^\dagger\alpha^{m-1}\mathcal{J}^{m-2}]\mathcal{A}^{m-2}[\mathcal{J}^{m-3}\alpha^{m-2}\mathbf{W}^{m-2}\mathbf{J}^{m-2}\beta^{m-1}\mathbf{W}^{m-1}]. \tag{3.11}$$

Now

$$\|\mathcal{J}^{m-2}\alpha^{m-1}\mathbf{W}^{m-1}\|^2 \leqslant \sup_\varphi \langle\varphi|\alpha^{m-1}(\mathcal{J}^{m-2})^\dagger\mathcal{J}^{m-2}\alpha^{m-1}|\varphi\rangle/\|\varphi\|^2$$

because $\|\mathbf{W}^m\|^2 = 1$. Here, $\sup_\varphi$ indicates the upper bound. But, we observe that

$$\begin{aligned} (\mathcal{J}^{m-2})^\dagger\mathcal{J}^{m-2} &= (\mathcal{Y}^{m-2})^\dagger\mathcal{Y}^{m-2} = \mathbf{K}(\Gamma^{m-2})^\dagger\Gamma^{m-2}\mathbf{K}^\dagger/(\beta^{m-2})^2 \\ &= \mathbf{K}(\Gamma^{m-2})^\dagger\,\mathrm{Im}(\Gamma^{m-2})^{-1}\Gamma^{m-2}\mathbf{K}^\dagger = -\mathrm{Im}\,\mathcal{F}(\Sigma^{m-2}) = -\mathrm{Im}\,\Sigma^{m-1}. \end{aligned} \tag{3.12}$$

The above result follows from equation (3.3*b*). Thus we have

$$\|\mathcal{J}^{m-2}\alpha^{m-1}\mathbf{W}^{m-1}\|^2 \leqslant \sup_\varphi \langle\varphi|\alpha^{m-1}(-\mathrm{Im}\,\Sigma^{m-1})\alpha^{m-1}|\varphi\rangle/\|\varphi\|^2.$$

We now define $|\chi\rangle = \alpha^{m-1}|\varphi\rangle$, and then we get

$$\|\mathcal{J}^{m-2}\alpha^{m-1}\mathbf{W}^{m-1}\|^2 \leqslant \sup_\chi \frac{\langle\chi|(-\mathrm{Im}\,\Sigma^{m-1})|\chi\rangle}{\langle\chi|(1/\alpha^{m-1})^2|\chi\rangle} \leqslant \sup_\chi \frac{\langle\chi|(-\mathrm{Im}\,\Sigma^{m-1})|\chi\rangle}{\langle\chi|\mathrm{Im}(\mathbf{G}^{m-1})^{-1}|\chi\rangle}. \tag{3.13}$$

Now from equations (3.4) and (3.5) we observe that

$$\mathrm{Im}\,\Sigma^{m-1} = \mathrm{Im}\,\zeta^{m-2} \qquad (\mathbf{G}^{m-1})^{-1} = (\mathbf{G}_{\mathrm{vc}})^{-1} - \zeta^{m-1}$$

where

$$\zeta^{m-2} = \mathbf{K}\Gamma^{m-2}\mathbf{K}^{\dagger}.$$

Thus, equation (3.13) readily gives us

$$\|\mathcal{J}^{m-2}\alpha^{m-1}\mathbf{W}^{m-1}\|^2 \leqslant \sup_{\chi} \frac{\langle\chi|(-\operatorname{Im}\zeta^{m-2})|\chi\rangle}{\langle\chi|\operatorname{Im}(\mathbf{G}_{vc})^{-1}|\chi\rangle + \langle\chi|(-\operatorname{Im}\zeta^{m-2})|\chi\rangle}.$$

Now, since $\operatorname{Im}(\mathbf{G}_{vc})^{-1} \geqslant a > 0$ and $-\operatorname{Im}\zeta = -\operatorname{Im}\boldsymbol{\Sigma} \geqslant b > 0$, the above expression becomes

$$\|\mathcal{J}^{m-2}\alpha^{m-1}\mathbf{W}^{m-1}\|^2 \leqslant b/(a+b)$$

with both a and $b > 0$. Thus we immediately get

$$\|\mathcal{J}^{m-2}\alpha^{m-1}\mathbf{W}^{m-1}\|^2 = \lambda_1 < 1.$$

Again, in an exactly similar manner, we can get

$$\|\mathbf{J}^{m-1}\beta^{m}\mathbf{W}^{m}\|^2 = \lambda_2 < 1.$$

Thus, from equation (3.11) we have

$$\|\mathcal{A}^m\| \leqslant \lambda^2\|\mathcal{A}^{m-2}\| \qquad \lambda = (\lambda_1\lambda_2)^{1/2} < 1 \Rightarrow \|\mathcal{A}^m\| \leqslant \lambda^m\|\mathcal{A}^0\| \tag{3.14}$$

and from (3.9) we get

$$\|\Delta\boldsymbol{\Sigma}^m\| \leqslant \|\mathcal{J}^{m-1}\|\,\|\mathcal{J}^{m-2}\|\,\|\mathcal{A}^{m-1}\|. \tag{3.15}$$

Since $(\mathcal{J}^m)^{\dagger}\mathcal{J}^m = -\operatorname{Im}\boldsymbol{\Sigma}^{m+1}$ (from equation (3.12)) and $\boldsymbol{\Sigma}^{m+1}$ is always bounded, if we start from a bounded $\boldsymbol{\Sigma}^0$ (see proposition 2) we have $\|\mathcal{J}^m\| \leqslant \eta$, where η is some finite number. The same is true for an initial bounded starting $\|\mathcal{A}^0\|$. Thus from (3.14) and (3.15) we finally get

$$\|\Delta\boldsymbol{\Sigma}^m\| \leqslant \eta^2\lambda^{m-1}\|\mathcal{A}^0\| \Rightarrow \|\boldsymbol{\Sigma}^p - \boldsymbol{\Sigma}^q\| \leqslant \eta^2\|\mathcal{A}^0\| \sum_{j=q}^{p-1} \lambda^j \to 0 \qquad \text{as } p, q \to \infty.$$

Thus $\boldsymbol{\Sigma}^m$ is a Cauchy sequence and, therefore, will converge to a unique value as $m \to \infty$.

4. Conclusions

We have shown in this paper that the CCPA, introduced through the ASF, preserves all the analytic properties of the Green function and the self-energy and, therefore, guarantees non-negative density of states. This approximation retains the translational invariance of the effective medium.

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