# Multiple scattering of classical waves in systems with liquidlike correlations: Formulation as a liquid-state theory

C. J. Walden
*Faculty of Mathematical Studies, University of Southampton, Southampton SO17 1BJ, United Kingdom*
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The multiple-scattering theory for wave propagation in a medium with spherical inclusions is examined for the case where these exhibit liquidlike correlations. By adopting graphical methods previously employed in the context of disordered tight-binding models, the determination of the ensemble-averaged amplitude (one-particle) Green function is reduced to the solution of two central equations. One of these is an operator analog of the Ornstein-Zernike (OZ) equation of liquid-state theory. The other describes the self-consistent determination of an effective single-scatterer $T$ matrix. This formalism leads naturally to the definition of a direct and a total propagator, the former being identified with what is generally termed the medium propagator. It is demonstrated that a number of existing theories may be derived as closure approximations to the (exact) pseudo-OZ equation. By generalizing previous treatments of a given (effective medium) approximation, it is then shown how the intensity (two-particle) Green function may be derived in a manner that ensures energy conservation.

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## I. INTRODUCTION

The propagation of classical waves in random media has attracted considerable attention recently [1,2]. Much of this interest has been motivated by the search for Anderson localization [3–5], especially in the case of light waves, where its realization promises a variety of novel technological applications [6]. From the point of view of establishing contact with theoretical predictions, Anderson localization of light has the advantage of being uncomplicated by competing mechanisms: in the electronic context electron-electron and electron-phonon interactions cannot be ignored [7]. On the other hand, while localization of electrons in low-energy band tails is in some senses ubiquitous, this (low-frequency) limit is accompanied by a vanishing of the scattering "potential" for light waves [8]. The opposite high-frequency or geometrical-optics limit also yields no localization, which is expected to occur in some intermediate-frequency window. This forces us to study a regime in which the wavelength may be comparable with the scale of structural correlations in the medium [8]. Any theory should therefore take adequate account of such structure. For discrete random media, which are of interest here, this information is contained in the $n$-particle distribution functions $\rho^{(n)}(\mathbf{R}_1,\ldots,\mathbf{R}_n)$, $n=1,\ldots,N$, where $N$ is the number of particles (i.e., scatterers) in the system. In general, the $\rho^{(n)}$ depend also on the orientation of the particles, but, for simplicity, we will assume each scatterer to be spherically symmetric.

In order to study localization, we require a theory for the *intensity* or two-particle Green function, averaged over allowed configurations of scatterers. In particular, we should exclude cases where the scatterers overlap. Clearly this constraint introduces complications and, in order to make some headway, many authors [9–12] have considered point-scatterer models, for which positional correlations are absent. Such models allow one to study what may be termed dependent scattering effects, i.e., how the scattering efficiency of a given particle is modified by multiple scattering from the other particles. The net effect is generally an increase in the mean free path associated with wave propagation, above that predicted by a single-scattering treatment. Unfortunately, correlations between the scatterers give rise to similar effects, as demonstrated by the correlation-corrected single-scattering calculations of Saulnier *et al.* [13]. What is required is a formalism that treats both multiple scattering and correlations on an equal footing. The present paper provides one such formalism. In another paper we shall go on to exploit this in the case of isotropic scatterers. In particular, we will show how liquid-state methods may be used rather directly in obtaining the one-particle Green function. This yields an explicit solution for a simple (step-function) model of pair correlations and numerical results for more realistic correlation functions.

A large part of what appears here concerns the calculation of the averaged *amplitude* Green function in media whose constituent particles exhibit correlations of a liquidlike nature. The types of system we have in mind are colloidal suspensions or similar systems in which the liquidlike structure has been quenched in by, for example, some kind of gelation process. Whilst the amplitude Green function does not, in principle, yield information about localization, it forms an essential ingredient in any theory for the intensity Green function.

A great deal of insight has been gained in the study of disordered conductors by appealing to the predictions of tight-binding (TB) models [14]. The applicability of such an approach for classical waves is still far from clear, the principal difficulty being that, for the latter, the physi-

 

cally relevant regime corresponds to the positive-energy continuum [8]. Hence there are no true bound states to associate with each particle, only resonances.

Recent classical-wave band structure calculations [15,16] have established some correspondences between such single-scatterer resonances and the positions of band gaps, at least in the case of scalar waves [17]. For low packing fractions $\eta$ of scatterers the midgap frequencies coincide with the resonances. However, as $\eta$ is increased the gaps tend to move between the resonances. Datta *et al.* [15] suggest that, for small $\eta$, wave propagation is predominantly through the host medium, with the scatterers serving to impede the motion. For large $\eta$ one may visualize propagation in terms of a hopping between overlapping local resonant states.

This picture has considerable appeal, although at present it is not clear how one may put it to work in the form of a TB model. Based on the intuition gained from studying electronic systems, one would expect such an approach to be valid for narrow bands with wide gaps. At present it seems that this requirement is most likely to be met in acoustic systems, for which one may tailor both the velocity and impedance mismatch [16] between host and scatterer.

In the absence of any obvious prescription for interpreting the results of TB calculations in the context of classical-wave systems, let us choose instead a multiple-scattering approach. Fortunately, for the liquidlike systems that concern us here, many of the diagrammatic methods employed for disordered TB models may be carried over. In particular, the multiple-scattering formalism for the configurationally averaged amplitude Green function may be recast in a form that resembles a liquid-state theory. Logan and Winn [18] established an equivalent reformulation for a disordered single-band TB model and later extended this to multiple-band systems [19]. Their analysis in turn employed methods developed by Wertheim [20] for determining the dielectric constant of a nonpolar, polarizable fluid. The relation between this latter problem and multiple-scattering theory has not gone unnoticed in the literature [21]. However, the utility of a liquid-state theoretic approach appears not to have been explored before.

In this paper it will be shown that the Logan-Winn formalism leads naturally to theories for the amplitude Green function (e.g., the effective-medium approximation (EMA) of Roth [22,23]) for which the self-consistent equations involve only "on-shell" quantities. The formalism presented here is developed for classical scalar waves. Its generalization to the case of electromagnetic waves is straightforward, but requires some extra bookkeeping. The reader is referred to the paper by Davis and Schwartz [21] that discusses the quasicrystalline approximation (QCA) [24,25] and the EMA [22,23,26] for light waves in a notation that closely resembles the present one.

The remainder of the paper is arranged as follows. Section II sets up the multiple-scattering theory for the amplitude Green function in terms of the medium path operator. It is this quantity that corresponds to the average Green function in the TB language. Hence the topological analysis of the diagrams contributing to this may be achieved by direct translation from the paper by Logan and Winn [18]. However, the fact that we are now dealing with operators means that some additional work needs to be done to put the equations in a form suitable for calculation. In an angular momentum representation the central equations (which must be solved self-consistently) are shown to be reducible to an on-shell form. Off-shell quantities are not involved in the self-consistency and may be calculated once their on-shell counterparts have been found.

The above decoupling of the on-shell terms relies on the satisfaction of a nonoverlapping condition [27–29]. Section III shows how the Logan-Winn formalism, in which approximations are introduced as closures of an Ornstein-Zernike-like integral equation [30], may be used to guarantee this.

Section IV considers how to extend these ideas to the calculation of the intensity or two-particle Green function. Roth and Singh [31] and also Itoh *et al.* [32–34] have employed a "variational derivative" approach that allows this quantity to be determined, given its one-particle counterpart. Their treatments were for two specific theories of the amplitude Green function, viz., the quasicrystalline approximation with coherent potential (QCACP) [35,36] and the EMA. Using the present formalism, an analogous approach is applied, valid within any "closure" approximation for the amplitude Green function. Of particular significance for classical waves is that the energy conservation law differs from that for Schrödinger waves. It is verified for the EMA and a slightly more elaborate approximation, which includes repeated scattering between pairs of scatterers, that the above scheme generates theories that are consistent with this conservation law.

Finally, Sec. V provides a brief summary and draws some conclusions.

## II. MULTIPLE-SCATTERING FORMALISM

This section provides a formal analysis of the multiple-scattering theory for the amplitude Green function, averaged over the ensemble of possible configurations of the scatterers. By selective resummation of certain classes of diagrams it is shown how this may be rewritten to resemble a liquid-state theory. This leads to a natural distinction between a *direct* and a *total* propagator. It also yields an expression for the renormalized (or loop-corrected) $T$ matrix of a single scatterer, the form of which, it will be argued, should be preserved in developing new approximations. This feature is shared with the treatment of Lloyd [27], although his expression involves the bare reaction matrix rather than the $T$ matrix.

Consider a system of identical discrete spherical scatterers of dielectric constant $\epsilon(\omega)\epsilon_0(\omega)$ embedded in a host medium characterized by $\epsilon_0(\omega)$. For a given configuration we may write, for the (unaveraged) Green function (with a dependence on frequency $\omega$ understood),

$$\mathcal{G}(\mathbf{r},\mathbf{r}') = G_0(\mathbf{r}-\mathbf{r}') + \int d\mathbf{r}_1 d\mathbf{r}_2 G_0(\mathbf{r}-\mathbf{r}_1)\mathcal{T}(\mathbf{r}_1,\mathbf{r}_2)G_0(\mathbf{r}_2-\mathbf{r}'), \tag{1}$$

which serves to define $\mathcal{T}(\mathbf{r}_1,\mathbf{r}_2)$ the random $T$ matrix for the whole system. The host Green function $G_0(\mathbf{r}-\mathbf{r}')$ satisfies

$$\left[\nabla^2 + k^2\right] G_0(\mathbf{r}-\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}'), \tag{2}$$

with the wave number $k$ in the host medium given by $k^2 = \epsilon_0(\omega)(\omega^2/c^2)$, $c$ being the free-space phase velocity.

The spirit of the multiple-scattering approach is to expand $\mathcal{T}(\mathbf{r}_1,\mathbf{r}_2)$ in a series of terms representing single scattering, double scattering, etc. For this purpose it is convenient to employ an operator notation, in terms of which Eq. (1) becomes

$$\hat{\mathcal{G}} = \hat{G}_0 + \hat{G}_0\hat{\mathcal{T}}\hat{G}_0. \tag{3}$$

$\mathcal{G}(\mathbf{r},\mathbf{r}')$ may now be identified with the matrix element $\langle \mathbf{r} \mid \hat{\mathcal{G}} \mid \mathbf{r}' \rangle$ and so on.

Consider now the scattering path operator [37] $\hat{\mathcal{T}}_{\alpha\beta}$. This acts on the wave incident at scatterer $\beta$ (located at $\mathbf{R}_\beta$) and provides the wave scattered from scatterer $\alpha$, incorporating all intermediate (multiple) scattering. It may be expanded as

$$\hat{\mathcal{T}}_{\alpha\beta} = \hat{t}_\alpha\delta_{\alpha\beta} + (1-\delta_{\alpha\beta})\hat{t}_\alpha\hat{G}_0\hat{t}_\beta + \sum_{\substack{\gamma\neq\alpha\\ \gamma\neq\beta}} \hat{t}_\alpha\hat{G}_0\hat{t}_\gamma\hat{G}_0\hat{t}_\beta + \cdots, \tag{4}$$

where $\hat{t}_\alpha \equiv \hat{t}(\mathbf{R}_\alpha)$ are (bare) single-scatterer $T$ matrices. Each describes the scattering from an isolated scatterer embedded at position $\mathbf{R}_\alpha$ in the host medium. Let us assume, for the moment, that these are known quantities. In fact, for many purposes it suffices to know only their "on-shell" parts, which may be calculated via standard phase-shift methods [38,39]. The total $T$ matrix $\hat{\mathcal{T}}$ is obtained from $\hat{\mathcal{T}}_{\alpha\beta}$ by summing over all possible initial and final scatterers $\alpha,\beta$.

For the ensemble-averaged Green function $\hat{G} \equiv \langle\hat{\mathcal{G}}\rangle$ we may adopt a similar approach:

$$\hat{G} = \hat{G}_0 + \hat{G}_0\hat{T}\hat{G}_0. \tag{5}$$

The average total $T$ matrix may be expressed as

$$\hat{T} \equiv \langle\hat{\mathcal{T}}\rangle = \int d\mathbf{R}d\mathbf{R}'\hat{\tau}(\mathbf{R}-\mathbf{R}'), \tag{6}$$

with the medium path operator [23] $\hat{\tau}(\mathbf{R}-\mathbf{R}')$ defined via

$$\hat{\tau}(\mathbf{R}-\mathbf{R}') = \left\langle \sum_{\alpha,\beta} \delta(\mathbf{R}-\mathbf{R}_\alpha)\hat{\mathcal{T}}_{\alpha\beta}\delta(\mathbf{R}'-\mathbf{R}_\beta) \right\rangle. \tag{7}$$

Before proceeding further, let us make precise what is meant by the ensemble average of some function $f(\mathbf{R}_1,\ldots,\mathbf{R}_n)$ of $n \leq N$ of the positions of the particles in the system. This is defined via

$$\langle f\rangle = Z_N^{-1}\int\cdots\int d\mathbf{R}_1\cdots d\mathbf{R}_N f e^{-\Phi_N(\mathbf{R}_1,\ldots,\mathbf{R}_N)/k_BT}, \tag{8}$$

where $T$ refers here to the temperature, $k_B$ is Boltzmann's constant and the configurational integral $Z_N$ is given by

$$Z_N = \int\cdots\int d\mathbf{R}_1\cdots d\mathbf{R}_N e^{-\Phi_N(\mathbf{R}_1,\ldots,\mathbf{R}_N)/k_BT}. \tag{9}$$

Here the $N$ particles of the system are assumed to interact via a potential $\Phi_N(\mathbf{R}_1,\ldots,\mathbf{R}_N)$. While it is often convenient to approximate this by a sum of pairwise additive potentials, this is not a necessary restriction as regards the present formalism. The following discussion will focus on the limit $N\to\infty$, $V\to\infty$, where $V$ is the volume of the system, such that $N/V = \rho$, a constant number density of scatterers. This, together with the assumed absence of an external potential, leads to a macroscopically homogeneous system and permits us to write $\mathbf{R}-\mathbf{R}'$ as the argument to the medium path operator.

A multiple-scattering expansion of $\hat{\tau}(\mathbf{R}-\mathbf{R}')$ involves the distribution functions

$$\rho^{(n)}(\mathbf{R},\mathbf{R}_1,\ldots,\mathbf{R}_{n-2},\mathbf{R}') \equiv \frac{N!}{(N-n)!} \times \left\langle \sum_{\alpha_1}'\cdots\sum_{\alpha_n}' \delta(\mathbf{R}-\mathbf{R}_{\alpha_1})\cdots\delta(\mathbf{R}'-\mathbf{R}_{\alpha_n}) \right\rangle, \quad n = 1,\ldots,N, \tag{10}$$

where the primes on the summation indicate that no two indices $\alpha_i$ are equal. Note that in the limit $N\to\infty$ we may replace $N!/(N-n)!$ by unity.

The structure of this series becomes clear if we adopt a diagrammatic approach. Significant in this treatment is the fact that the corresponding graphs are *composite*, comprising a part that describes the multiple-scattering path (represented below by a continuous "chain") and a part that reflects the scatterer correlation. The topological analysis of such graphs has been considered previously by Wertheim [20] in the context of determining the dielectric constant of a nonpolar fluid. These ideas were subsequently employed by Logan and Winn [18] to study the density of states of a disordered tight-binding model. The adoption of such methods in the present case simply requires the identification of a set of rules for interpreting the diagrams. Since the topological arguments are well documented, their description here will be reasonably brief.

At this point it is helpful to separate $\hat{\tau}(\mathbf{R}-\mathbf{R}')$ into diagonal and off-diagonal parts

$$\hat{\tau}(\mathbf{R}-\mathbf{R}') = \rho\hat{t}^{(m)}(\mathbf{R})\delta(\mathbf{R}-\mathbf{R}') + \hat{\tau}_{\mathrm{OD}}(\mathbf{R}-\mathbf{R}'), \tag{11}$$

thereby defining a renormalized single-scatterer $T$ matrix $\hat{t}^{(m)}(\mathbf{R})$,

$$\rho\hat{t}^{(m)}(\mathbf{R}) = \left\langle \sum_{\alpha} \delta(\mathbf{R}-\mathbf{R}_\alpha)\hat{\mathcal{T}}_{\alpha\alpha} \right\rangle, \tag{12a}$$

$$\hat{\tau}_{\mathrm{OD}}(\mathbf{R}-\mathbf{R}')$$

$$= \left\langle \sum_{\alpha,\beta} \delta(\mathbf{R}-\mathbf{R}_\alpha)\hat{\mathcal{T}}_{\alpha\beta}(1-\delta_{\alpha\beta})\delta(\mathbf{R}'-\mathbf{R}_\beta) \right\rangle. \tag{12b}$$

For the sake of clarity, in what follows the shorthand $\mathbf{R}_\alpha \equiv \alpha$, $d\mathbf{R}_\alpha \equiv d(\alpha)$ will be used.

Consider now the multiple-scattering series for $\hat{\tau}_{\mathrm{OD}}(\mathbf{R}_1-\mathbf{R}_2) \equiv \hat{\tau}_{\mathrm{OD}}(1,2)$. For each term of $\hat{\mathcal{T}}_{\alpha\beta}$ that involves $s$ distinct scatterers the averaging procedure introduces a factor of $\rho^{(s)}(1,2,\ldots,s) \equiv \rho^s g_s(1,2,\ldots,s)$, where $g_s$ is the normalized $s$-particle distribution function. It follows that we may write

$$\hat{\tau}_{\mathrm{OD}}(1,2) = \rho^2 g_2(1,2)\hat{\tau}^{(2)}(1,2) + \sum_{s=3}^{\infty} \int \rho^s g_s(1,2,\ldots,s) \times \hat{\tau}^{(s)}(1,2,\ldots,s)d(3)\cdots d(s), \tag{13}$$

where $\hat{\tau}^{(s)}(1,2,\ldots,s)$ is the sum of all terms appearing in $\hat{\mathcal{T}}_{12}$ that involve $s$ scattering centers. The graph-theoretical analysis of $\hat{\tau}_{\mathrm{OD}}(1,2)$ now parallels that of Wertheim [20] and Logan and Winn [18]. The essential difference between these papers and the present treatment is that we are now considering operators that act on the wave coordinates. It is this feature that makes it convenient to work with the scattering path operator rather than directly with the Green function $\hat{\mathcal{G}}$.

Central to the aforementioned analysis is the result from liquid-state graph theory that

$$g_s(1,\ldots,s) = \prod_{t=2}^{s} \prod_{\text{all } Q_t} [1+\gamma_t(Q_t)]. \tag{14}$$

Here $Q_t$ refers to a particular combination of $t$ points, chosen from $1,2,\ldots,s$, and $\gamma_t(Q_t)$ are so-called direct connectors to be described below. Stell [40] provides a derivation of Eq. (14) for a pairwise additive potential

$$\Phi_N(1,\ldots,N) = \sum_{1\le\alpha<\beta\le N} \phi_2(\alpha,\beta). \tag{15}$$

In fact, his treatment remains valid even in the presence of an external potential $V_{\mathrm{ext}} = \sum_{1\le\alpha\le N}\phi_1(\alpha)$, at the cost of introducing a position-dependent singlet density $\rho^{(1)}(\mathbf{R}) = \langle\sum_\alpha \delta(\mathbf{R}-\mathbf{R}_\alpha)\rangle$.

In the discussion that follows it is helpful to bear in mind a number of concepts from graph theory. For further details the reader is referred to Wertheim [20], Stell [40], and Mayer and Mayer [41]. The points appearing in a given graph fall into two categories: root points and field points, the latter representing coordinates that are integrated over. A one-articulation point (1AP) is defined as a point $\alpha$ which, when removed, causes the graph to separate into two or more pieces, at least one of which contains no root points. Each graph of field points so detached is termed a one-articulated subgraph (1ASG). In what follows the term *simple* will be used to denote a graph in which, between every pair of adjacent points, there is only one path that consists of a single bond. As mentioned above, the graphs appearing in $\hat{\tau}_{\mathrm{OD}}(1,2)$ are *composite* since they involve bonds relating to the correlation functions together with the propagators of the multiple-scattering series.

We will not consider in any detail the construction of $g_s(1,\ldots,s)$ from more fundamental constituents since this is the domain of liquid-state theory. However, it does seem desirable to clarify the meaning of the direct connectors $\gamma_t(Q_t)$. For a potential of the form given in (15), $\gamma_t(Q_t)$ is defined as the sum of all distinct simple graphs with $t$ root points labeled by members of the set $Q_t$ and at least one field point. These are connected via Mayer $f$ bonds

$$f(\alpha,\beta) = e^{-\phi_2(\alpha,\beta)/k_BT} - 1 \tag{16}$$

such that every graph is free of 1APs. In addition, every field point is connected to every root point by at least one path that does not pass through any intermediate root points. Also, there is no single-bond path between any pair of root points.

As Wertheim [20] points out, Eq. (14) is valid for potentials that include $n$-body terms, with $n \ge 3$. In that case the direct connectors $\gamma_t(Q_t)$ will involve $n$-body generalizations of the $f$ bond (see the work of Mayer and Mayer [41]). Rather than dwell on the details of these correlation graphs, let us assume henceforth that the $\gamma_t$ are known.

Turning now to the multiple-scattering graphs, the first few (chain) diagrams contributing to $\hat{\tau}^{(s)}(1,\ldots,s)$ for $s = 2,\ldots,5$ are given in Fig. 1. They correspond to what Frisch [42] terms "half-dressed" diagrams (see Fig. 2), where repeated visits of the multiple-scattering chain to a given site are represented by "links." In his notation, "fully-dressed" diagrams are formed by the incorporation of correlation connectors. In the expansion of the self-energy, these latter quantities are constructed from short-ranged correlation functions $h_s(1,\ldots,s)$.

Rather than repeat the formal analysis of $\hat{\tau}_{\mathrm{OD}}(1,2)$, which parallels that for the quantity $\mathbf{A}(\mathbf{R},\mathbf{R}')$ in Wertheim's paper [20], it seems more helpful to provide some examples. Consider the graph of Fig. 2(a). Associated with this is a factor $\rho^3 g_3(1,2,3)$. Now by Eq. (14) we may write

$$\begin{aligned} g_3(1,2,3) &= g_2(1,2)g_2(1,3)g_2(2,3)[1+\gamma_3(1,2,3)] \\ &= g_2(1,2)g_2(1,3) + g_2(1,2)g_2(1,3)h_2(2,3) \\ &\quad + g_2(1,2)g_2(1,3)g_2(2,3)\gamma_3(1,2,3), \end{aligned} \tag{17}$$

where $h_2(2,3) = g_2(2,3) - 1 = \gamma_2(2,3)$. If we take the first term on the right-hand side of this expression it is

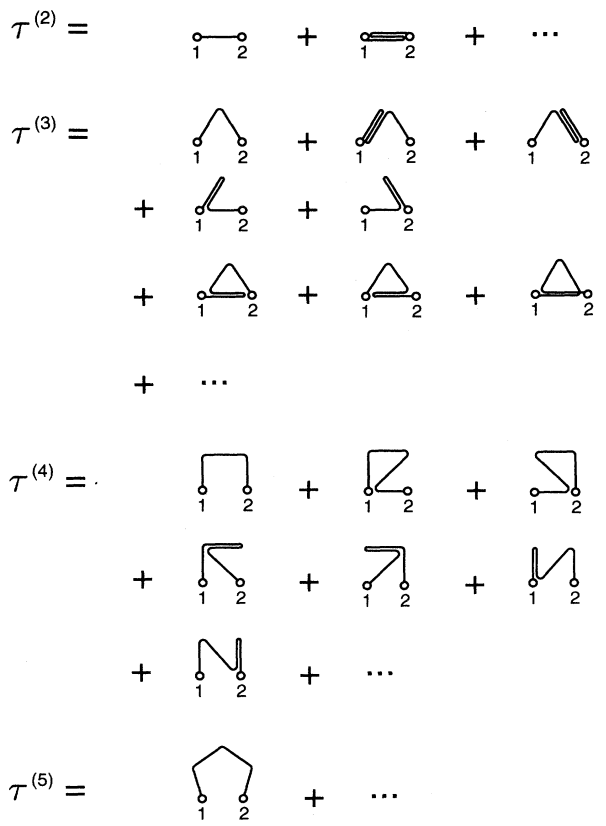

FIG. 1. Graphs appearing in $\hat{\tau}^{(s)}(1,\ldots,s)$ for $s=2,\ldots,5$. The rules for interpreting the diagrams are $\hat{t}(\alpha)$ for each *stage*, i.e., each association of a chain with a given point $\alpha$, and $\hat{G}_0$ for each portion of chain (bond) connecting two stages. Field points will be integrated over after multiplication by a suitable correlation graph.

easy to see that the associated graph (Fig. 3) is reducible via a suitable cut at 1. This separates the graph into two pieces since there is no direct $\gamma$ connector between points 2 and 3. The fragment containing points 1 and 3 involves a scattering path (chain) that begins and ends on the same scatterer, 1. It therefore contributes to the renormalized $T$ matrix $\hat{t}^{(m)}(1)$.

We may employ a similar analysis with the graph in Fig. 4. With the aid of Eq. (14) it is possible to identify a term $g_2(1,2)g_3(1,3,4)$ in the expression for $g_4(1,2,3,4)$. The associated $\hat{\tau}_{\rm OD}(1,2)$ graph is seen to be reducible via a maximal cut at 1, i.e., a cut that leaves the largest

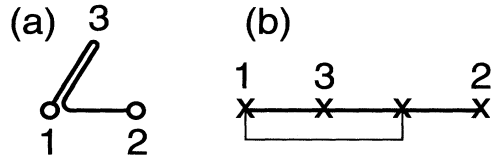

FIG. 2. (a) Typical chain graph appearing in $\hat{\tau}^{(3)}(1,2,3)$ and (b) equivalent "half-dressed" diagram in a notation derived from that of Frisch [42].

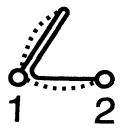

FIG. 3. Reducible graph contributing to $\hat{\tau}_{\rm OD}(1,2)$. The dotted lines represent $g_2$ bonds.

possible subgraph attached to root point 1. Once again this subgraph gives a contribution to $\hat{t}^{(m)}(1)$. The remaining terms all involve direct connectors between 2 and members of the set of points $(1,3,4)$, which ensure that the corresponding graphs appearing in $\hat{\tau}_{\rm OD}(1,2)$ are irreducible.

In general we may identify 1ASG decorations of each point, which may all be summed to give a renormalized $T$ matrix. In this way we may form $\hat{\tau}_{\rm OD}(1,2)$ from graphs free from 1APs, but with each stage contributing $\hat{t}^{(m)}$ rather than $\hat{t}$. Now let us define a one-chain bridge point (1BP) as a field point whose removal causes the graph to fall into two pieces, one attached to each root point. This allows us to identify a subset of graphs which are free from 1BPs, whose sum is defined by $\rho\hat{t}^{(m)}(\alpha)\hat{C}(\alpha,\beta)\rho\hat{t}^{(m)}(\beta)$. Such graphs will be termed strongly irreducible.

Clearly, we may use the same chain graphs as Fig. 1 in constructing $\hat{C}(1,2)$ as long as we slightly modify the rules for interpreting them. In particular, each interior stage now contributes $\hat{t}^{(m)}$ and each end stage yields 1. Also a factor of $\rho$ accompanies each field point. Associated with each such graph is a sum of correlation graphs, chosen to render the composite graph strongly irreducible.

Having identified $\hat{C}(1,2)$, it follows that $\hat{\tau}_{\rm OD}(1,2)$ may be written in the form

$$\hat{\tau}_{\rm OD}(1,2)=\rho\hat{t}^{(m)}(1)\hat{H}(1,2)\rho\hat{t}^{(m)}(2), \tag{18}$$

where

$$\hat{H}(1,2)=\hat{C}(1,2)+\int\hat{C}(1,3)\rho\hat{t}^{(m)}(3)\hat{H}(3,2)d(3). \tag{19}$$

The similarity between (19) and the Ornstein-Zernike (OZ) equation [30] of liquid-state theory suggests that $\hat{C}(\alpha,\beta)$ and $\hat{H}(\alpha,\beta)$ be termed *direct* and *total* propagators, respectively. In fact, $\hat{C}(\alpha,\beta)$ may be identified with the medium propagator discussed by a number of authors [27,43,21,23].

An analysis of the diagrams appearing in the definition

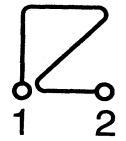

FIG. 4. Chain graph contributing to $\hat{\tau}^{(4)}(1,2,3,4)$.

of $\hat{t}^{(m)}(\alpha)$ reveals that it satisfies

$$\hat{t}^{(m)}(\alpha) = \hat{t}(\alpha) + \hat{t}^{(m)}(\alpha) \int \hat{H}(\alpha,\beta)\rho\hat{t}^{(m)}(\beta)\hat{G}_0 d(\beta)\hat{t}(\alpha). \tag{20}$$

In practical implementations of this equation $\hat{G}_0$ is generally expanded about the scattering centers $\alpha$ and $\beta$ (see below).

The operator equations (19) and (20) are exact and in the present formalism constitute the central equations of multiple-scattering theory. Let us consider how they may be put in a form suitable for calculation.

For the systems of interest, the scatterers are identical impenetrable spheres and so their closest distance of approach is $d$, the hard-sphere diameter. The implications of this are well known (see, e.g., the work of Lloyd and Berry [43]). In particular, it allows an effective refractive index to be calculated using only the on-shell part of the $T$ matrices. In another context, viz., muffin-tin models of liquid metals, it allows a similar on-shell evaluation of the density of states via the so-called Lloyd formula [44,27,45].

For the bare $T$ matrix, the on-shell part is given (in an angular momentum representation) by

$$\begin{aligned} t_{LL'}(k,k) &= t_l(k)\delta_{LL'} \\ &= \int d\mathbf{r} \int d\mathbf{r}' j_l(kr) Y_L^*(\hat{\mathbf{r}}) t(\mathbf{r},\mathbf{r}') j_{l'}(kr') Y_{L'}(\hat{\mathbf{r}}'), \end{aligned} \tag{21}$$

where $\mathbf{r}$ and $\mathbf{r}'$ are coordinates relative to the center of the scatterer, $j_l(kr)$ are spherical Bessel functions, and $Y_L(\hat{\mathbf{r}})$ are spherical harmonics, $L \equiv (l,m)$ being a composite angular momentum label. A similar equation defines the on-shell part of $\hat{t}^{(m)}$. The significant feature from a computational viewpoint is that $t_l(k)$ may be determined by standard phase-shift methods [38,39].

Consider now Eq. (20) for $\hat{t}^{(m)}(\alpha)$. Provided the scatterers at $\mathbf{R}_\alpha$ and $\mathbf{R}_\beta$ do not overlap we can expand $\hat{G}_0$ as

$$\begin{aligned} \langle \mathbf{r} \mid \hat{G}_0 \mid \mathbf{r}' \rangle &= G_0(\mathbf{r},\mathbf{r}') \\ &= \sum_{L,L'} j_l(kr_\alpha) Y_L(\hat{\mathbf{r}}_\alpha) \\ &\quad \times G^0_{LL'}(\mathbf{R}_\alpha - \mathbf{R}_\beta) j_{l'}(kr'_\beta) Y^*_{L'}(\hat{\mathbf{r}}'_\beta), \end{aligned} \tag{22}$$

where $\mathbf{r}_\alpha \equiv \mathbf{r} - \mathbf{R}_\alpha$, $\mathbf{r}'_\beta \equiv \mathbf{r}' - \mathbf{R}_\beta$ (see, e.g., [45]). Here

$$\begin{aligned} &G^0_{LL'}(\mathbf{R}_\alpha - \mathbf{R}_\beta) \\ &= -4\pi i k \sum_{L''} i^{l''} C^{L''}_{LL'} h^+_{l''}(k \mid \mathbf{R}_\alpha - \mathbf{R}_\beta \mid) Y_{L''}(\widehat{\mathbf{R}_\alpha - \mathbf{R}_\beta}), \end{aligned} \tag{23}$$

$h^+_{l''}(k|\mathbf{R}_\alpha - \mathbf{R}_\beta|)$ being (outgoing) spherical Hankel functions and $C^{L''}_{LL'}$ Gaunt numbers, defined by

$$C^{L''}_{LL'} = \int Y_L(\hat{\mathbf{x}}) Y^*_{L''}(\hat{\mathbf{x}}) Y_{L'}(\hat{\mathbf{x}}) d\Omega_{\mathbf{x}}, \tag{24}$$

where the integral is over all solid angles.

Writing now

$$\begin{aligned} &\langle \mathbf{r} \mid \hat{H}(\alpha,\beta) \mid \mathbf{r}' \rangle \\ &= \sum_{L,L'} j_l(kr_\alpha) Y_L(\hat{\mathbf{r}}_\alpha) H_{LL'}(\alpha,\beta) j_{l'}(kr'_\beta) Y^*_{L'}(\hat{\mathbf{r}}'_\beta) \end{aligned} \tag{25}$$

and

$$\begin{aligned} &\langle \mathbf{r} \mid \hat{C}(\alpha,\beta) \mid \mathbf{r}' \rangle \\ &= \sum_{L,L'} j_l(kr_\alpha) Y_L(\hat{\mathbf{r}}_\alpha) C_{LL'}(\alpha,\beta) j_{l'}(kr'_\beta) Y^*_{L'}(\hat{\mathbf{r}}'_\beta), \end{aligned} \tag{26}$$

which serves to define $H_{LL'}(\alpha,\beta)$ and $C_{LL'}(\alpha,\beta)$, it is seen that

$$\begin{aligned} t^{(m)}_{LL'}(k,k) &= t^{(m)}_l(k)\delta_{LL'}, \\ t^{(m)}_l(k) &= t_l(k) + \rho \sum_{L_1} \int t^{(m)}_l(k) H_{LL_1}(\alpha,\beta) t^{(m)}_{l_1}(k) \\ &\quad \times G^0_{L_1 L}(\beta,\alpha) d(\beta) t_l(k) \end{aligned} \tag{27}$$

and

$$\begin{aligned} H_{LL'}(\alpha,\beta) &= C_{LL'}(\alpha,\beta) + \sum_{L''} \rho t^{(m)}_{L''}(k) \\ &\quad \times \int C_{LL''}(\alpha,\gamma) H_{L''L'}(\gamma,\beta) d(\gamma). \end{aligned} \tag{28}$$

The OZ form (28) is reminiscent of that for a fluid mixture, with components labeled by $L$, etc. Winn and Logan [19] derived a similar equation for a multiple-band TB model. For the case of a Yukawa hopping matrix they were able to further exploit this analogy and obtain, for the EMA, a solution in closed form. Unfortunately, in the present case this approach is hampered by the fact that $C_{LL'}(\alpha,\beta)$ and $H_{LL'}(\alpha,\beta)$ depend on the vector $\mathbf{R}_\alpha - \mathbf{R}_\beta$, rather than just its modulus. In general, therefore, the solution of (28) requires rather involved techniques [46–48]. For isotropic scatterers, however, such a vectorial dependence does not appear and a "one-component" OZ equation obtains. A solution of this problem, which exploits the liquid-state analogy, will be presented elsewhere [49].

Note that Eqs. (27) and (28) involve only on-shell $T$ matrices. This result is a consequence of the nonoverlapping condition imposed on the scatterers, which implies

$$\hat{H}(\alpha,\beta) = 0 \quad \text{for } |\mathbf{R}_\alpha - \mathbf{R}_\beta| < d. \tag{29}$$

While this is clearly true for any exact treatment, which incorporates *all* strongly irreducible diagrams contributing to $\hat{C}(\alpha,\beta)$, it is not automatically satisfied in an approximate theory.

This problem, or its equivalent, was addressed by Lloyd [27], who pointed out the need for exercising care in choosing which terms to include in the series for $\hat{C}(\alpha,\beta)$. Singh and Roth [28] later pointed out an error in his

prescription. They also noted that other, well established, theories, such as the quasicrystalline approximation [24,25] and that due to Ishida and Yonezawa [50], were also deficient in this respect. We consider this issue in the following section.

## III. CLOSURE APPROXIMATIONS

On a practical level, evaluation of the above resummation scheme requires the introduction of approximations, not least because distribution functions $g_s(1,\ldots,s)$ generally are only available for $s \leq 2$. A variety of approximate multiple-scattering theories have been developed over the years, via a number of routes. It is instructive to consider how they relate to the present (OZ) formalism. In particular, it will be shown that several of these may be described in an extremely compact fashion as closure approximations to the pseudo-OZ equation (19), although in most cases these are supplemented by independent approximations for the renormalized $T$ matrix $\hat{t}^{(m)}$. Logan and Winn [18] have already established such a connection in a tight-binding language and concluded that the EMA, in the guise of the single-superchain approximation [20], provides the simplest approximation that is consistent with Eqs. (19) and (20). Before focusing on this and arguing the merits of Logan and Winn's strategy in developing systematic improvements to the EMA, we examine some of the aforementioned multiple-scattering theories.

Roth [23] has provided a summary of a number of theories that have the structure

$$\hat{\tau}(1,2) = \rho\hat{t}_c(1)\left[\delta(1,2) + \int \hat{G}^{(m)}(1,3)\hat{\tau}(3,2)d(3)\right], \tag{30}$$

where $\hat{G}^{(m)}(\alpha,\beta)$ is termed the medium propagator. Separating the diagonal and off-diagonal parts of the scattering path operators $\hat{\tau}(1,2)$ and $\hat{\tau}(3,2)$ [cf. Eq. (11)] we may show that

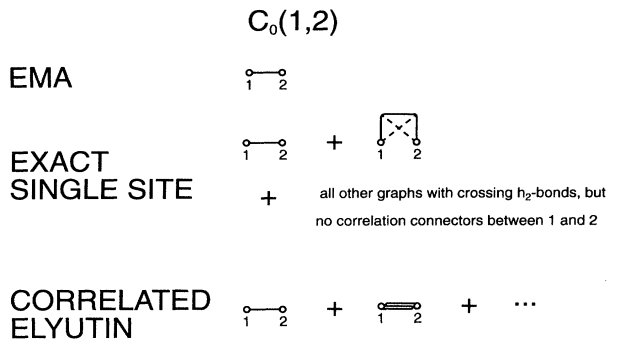


FIG. 5. Theories that satisfy the nonoverlapping condition and remain consistent with the OZ formalism.

$$\hat{\tau}_{\mathrm{OD}}(1,2) = \rho\hat{t}_c(1)\left[\hat{G}^{(m)}(1,2)\rho\hat{t}^{(m)}(2) + \int \hat{G}^{(m)}(1,3)\hat{\tau}_{\mathrm{OD}}(3,2)d(3)\right]. \tag{31}$$

Now, with the definition of $\hat{H}(\alpha,\beta)$ of Eq. (18) it is a simple matter to show that

$$\left[\hat{t}_c(1)\right]^{-1}\hat{t}^{(m)}(1)\hat{H}(1,2) = \hat{G}^{(m)}(1,2) + \int \hat{G}^{(m)}(1,3)\rho\hat{t}^{(m)}(3)\hat{H}(3,2)d(3). \tag{32}$$

This may be reduced to the OZ form (19) if we identify $\hat{t}_c(1)$ with $\hat{t}^{(m)}(1)$ and $\hat{G}^{(m)}(\alpha,\beta)$ with the direct propagator $\hat{C}(\alpha,\beta)$.

To check that this is internally consistent we consider the diagonal part $\rho\hat{t}^{(m)}(1)$ of $\hat{\tau}(1,2)$, which, by (30), should satisfy

$$\rho\hat{t}^{(m)}(1) = \rho\hat{t}_c(1)\left\{1 + \left[\hat{C}(1,1) + \int \hat{C}(1,3)\rho\hat{t}^{(m)}(3)\hat{H}(3,1)d(3)\right]\rho\hat{t}^{(m)}(1)\right\}, \tag{33}$$

Now, since $\hat{H}(\alpha,\beta)$ is, by definition, off diagonal in the scatterer indices, we have from the pseudo-OZ equation (19) that

$$0 = \hat{C}(1,1) + \int \hat{C}(1,3)\rho\hat{t}^{(m)}(3)\hat{H}(3,1)d(3), \tag{34}$$

so that our identification of $\hat{t}^{(m)}(1)$ and $\hat{t}_c(1)$ is indeed consistent.

In Roth's discussion, the renormalized $T$ matrix $t_c(\alpha)$ is expressed in terms of $\hat{v}_\alpha$, the scattering potential (operator) due to the particle $\alpha$,

$$\hat{t}_c(\alpha) = \hat{v}_\alpha + \hat{v}_\alpha\hat{G}_1(\alpha)\hat{t}_c(\alpha). \tag{35}$$

In the present context $\hat{v}_\alpha$ may be written

$$\hat{v}_\alpha = \epsilon_0(\omega)\frac{\omega^2}{c^2}[1-\epsilon(\omega)]\hat{\Theta}_\alpha, \tag{36}$$

where

$$\langle\mathbf{r}|\hat{\Theta}_\alpha|\mathbf{r}'\rangle = \delta(\mathbf{r}-\mathbf{r}')\Theta(a-|\mathbf{r}-\mathbf{R}_\alpha|), \tag{37}$$

with $a$ the radius of the scatterer and $\Theta(r)$ a Heaviside step function.

We note in passing that this depends on frequency $\omega$. In Schrödinger language this means that we are dealing with an energy-dependent potential. The prime consequence of this is that the system obeys a conservation law different from that usually encountered for electrons [51,52]. It is important to preserve this in formulating a theory for energy transport. This will be discussed in Sec. IV.

Defining now $\hat{G}'_1(\alpha) \equiv \hat{G}_1 - \hat{G}_0$ allows us to write

$$t_c(\alpha) = [\hat{v}_\alpha + \hat{v}_\alpha \hat{G}_0 \hat{v}_\alpha + \cdots][1 + \hat{G}'_1(\alpha)\hat{t}_c(\alpha)] = \hat{t}(\alpha) + \hat{t}(\alpha)\hat{G}'_1(\alpha)\hat{t}_c(\alpha), \quad (38)$$

where we identified the bare $T$ matrix

$$\hat{t}(\alpha) = \hat{v}_\alpha + \hat{v}_\alpha \hat{G}_0 \hat{v}_\alpha + \cdots = [1 - \hat{G}_0 \hat{v}_\alpha]^{-1} \hat{v}_\alpha. \quad (39)$$

This permits a more direct comparison to be made with the result (20), which implies that $\hat{G}'_1(\alpha)$ should take the form

$$\hat{G}'_1(\alpha) = \int \hat{H}(\alpha,\beta)\rho\hat{t}^{(m)}(\beta)\hat{G}_0 d(\beta). \quad (40)$$

Table I summarizes the theories discussed by Roth, viz., the QCA [24,25] and its self-consistent generalization (QCACP) [35,36], the self-consistent approximation of Schwartz and Ehrenreich [53] (SE), the theory of Ishida and Yonezawa [50] (IY), and the EMA [22,23].

Clearly the SE and EMA are closest in spirit to the OZ formalism in that they satisfy Eq. (40) and make approximations only to $\hat{C}(\alpha,\beta)$. The QCA makes perhaps the most drastic approximation for $\hat{G}'_1(\alpha)$, so that the $T$ matrix of a single scatterer is not renormalized. Both this and its coherent-potential generalization, which replaces $\hat{t}$ by the $T$ matrix of a scatterer in a self-consistently determined effective medium, are deficient with respect to their treatment of the local environment around a given scatterer. At sufficiently large packing fractions $\eta$, the presence of short-range order casts doubt on the applicability of a uniform effective medium that abuts on the boundary of the scatterer.

The rather cumbersome expression for $\hat{G}'_1(\alpha)$ in the IY approximation may be simplified in the following manner. First [18], let us relabel $\hat{H}(\beta,\gamma)$, which satisfies the pseudo-OZ equation (19) as $\hat{H}_m(\beta,\gamma)$. Now, by defining a new $\hat{H}(\beta,\gamma)$ via

$$\hat{H}(\beta,\gamma) = \hat{C}(\beta,\gamma) + g_2(\beta,\gamma)[\hat{H}_m(\beta,\gamma) - \hat{C}(\beta,\gamma)], \quad (41)$$

we arrive at an equation for $\hat{G}'_1(\alpha)$ that is consistent with (40). Notice that the expression (41) for $\hat{H}(\beta,\gamma)$, taken together with the IY definition for $\hat{C}(\beta,\gamma)$, incorporates an explicit factor of $g_2(\beta,\gamma)$. Hence, in the case of hard spheres, for which $g_2(\beta,\gamma) = 0$ when $|\mathbf{R}_\beta - \mathbf{R}_\gamma| < d$, the nonoverlapping condition (29) is seen to hold for this new $\hat{H}(\beta,\gamma)$. However, the theory is somewhat unsymmetrical since this modified quantity does not enter into the expression for $\hat{\tau}(\alpha,\beta)$, but rather $\hat{H}_m(\beta,\gamma)$, which does not satisfy the nonoverlapping condition. In the context of liquid metals this forces one to modify the formula for the density of states [28].

Recall that the nonoverlapping condition (29) is important in practical applications where the angular momentum decomposition of Eqs. (21)–(28) is generally employed. Of the above theories, the only one that meets this requirement while remaining fully consistent with the OZ formalism is the EMA. This may be seen most readily by recognizing that the integral appearing in the right-hand column of Table I may be identified as $\hat{H}(\alpha,\beta) - \hat{C}(\alpha,\beta)$. Hence

$$\hat{C}(\alpha,\beta) = g_2(\alpha,\beta)\hat{G}_0 + h_2(\alpha,\beta)[\hat{H}(\alpha,\beta) - \hat{C}(\alpha,\beta)]. \quad (42)$$

Rearranging this and using $g_2(\alpha,\beta) = h_2(\alpha,\beta) + 1$, we may identify an explicit factor of $g_2(\alpha,\beta)$.

This suggests that a way of amending a theory that fails to satisfy (29) is to multiply the existing expression for $\hat{H}(\alpha,\beta)$ by $g_2(\alpha,\beta)$. Such "energy-shell" extensions of the QCA and IY have been discussed previously by Singh and Roth [28] in connection with evaluating the density of states of a liquid metal via Lloyd's formula (see also Schwartz *et al.* [29]). However, theories of this form are no longer derivable within the OZ framework and there is evidence [28,21] that they fail to deal adequately with the effects of short-range order.

The EMA is widely regarded as one of the most promising approximations in this respect. It may be considered as the simplest in a class of theories described by closure approximations of the form

$$\hat{C}(\alpha,\beta) = g_2(\alpha,\beta)\hat{C}_0(\alpha,\beta) + h_2(\alpha,\beta)[\hat{H}(\alpha,\beta) - \hat{C}(\alpha,\beta)]. \quad (43)$$

All of these obey the nonoverlapping condition and are consistent with the OZ formalism. Figure 5 contains a summary of some approximations for $\hat{C}_0(\alpha,\beta)$. What we have termed the correlated Elyutin theory was introduced in a TB context by Winn and Logan [54] in an attempt to recover the correct low-density behavior of the density of states. A multiple-scattering equivalent

TABLE I. The propagators $\hat{G}'_1(\alpha)$ and $\hat{C}(\alpha,\beta) \equiv \hat{G}^{(m)}(\alpha,\beta)$ for various theories. Roth [23] uses the initials GKM of the original authors to denote the QCACP.

| Theory | $\hat{G}'_1(\alpha)$ | $\hat{C}(\alpha,\beta)$ |
|---|---|---|
| QCA | $0$ | $g_2(\alpha,\beta)\hat{G}_0$ |
| QCACP≡ GKM | $\hat{G} - \hat{G}_0$ | $\hat{G}_0 + h_2(\alpha,\beta)\hat{G}$ |
| SE | $\int \hat{H}(\alpha,\beta)\rho\hat{t}^{(m)}(\beta)\hat{G}_0 d(\beta)$ | $g_2(\alpha,\beta)\hat{G}_0 + h_2(\alpha,\beta)\int \hat{H}(\alpha,\gamma)\rho\hat{t}^{(m)}(\gamma)\hat{G}_0 d(\gamma)$ |
| IY | $\int \hat{C}(\alpha,\beta)\rho\hat{t}^{(m)}(\beta)\Big(\hat{G}_0 + \int \hat{H}(\beta,\gamma)\rho\hat{t}^{(m)}(\gamma)\hat{C}(\gamma,\alpha)d(\gamma)\Big)d(\beta)$ | $g_2(\alpha,\beta)\hat{G}_0$ |
| EMA | $\int \hat{H}(\alpha,\beta)\rho\hat{t}^{(m)}(\beta)\hat{G}_0 d(\beta)$ | $g_2(\alpha,\beta)\hat{G}_0 + h_2(\alpha,\beta)\int \hat{H}(\alpha,\gamma)\rho\hat{t}^{(m)}(\gamma)\hat{C}(\gamma,\beta)d(\gamma)$ |

to Elyutin's original theory [55] is obtained by setting $g_2(\alpha,\beta)=1$.

When substituted into (43) it is readily seen that the exact single-site theory (see Logan and Winn [18]) consists of (a) making the Kirkwood superposition approximation for all $g_s(1,\ldots,s)$ with $s>2$ and (b) retaining only single-site graphs in $\hat{C}(\alpha,\beta)$, i.e., folded chains representing recurrent scattering between a given pair of sites are omitted.

As indicated above, recurrent scattering terms, such as those incorporated in the correlated Elyutin theory, are important in the tight-binding context for describing the behavior at low density. In particular, it is in such a limit that one expects to observe a metal-insulator transition. In contrast, for classical waves, unless one has extremely strong scatterers (see, e.g., the work of van Tiggelen *et al.* [9,10,12]), an onset of localization seems more likely to occur at higher packing fractions. In this regime the EMA is probably adequate as far as amplitude properties are concerned. However, its extension to the calculation of intensity properties does not incorporate the maximally crossed diagrams, which are expected to be important for describing localization. Such diagrams do arise from an energy-conserving description of the intensity Green function based on the correlated Elyutin theory.

## IV. THE INTENSITY GREEN FUNCTION

In the previous sections we have focused on the determination of the average amplitude Green function $\hat{G}$. This yields information about the coherent wave, such as the effective refractive index and scattering mean free path, which may be extracted from its behavior in the long distance limit. As mentioned in the Introduction, it is more meaningful, at least as regards questions of localization, to consider the transport mean free path, for which a knowledge of the two-particle or intensity Green function $\langle\hat{\mathcal{G}}(z)\hat{\mathcal{G}}(z')\rangle$ is indispensible. Here $z$ and $z'$ denote complex frequencies, with infinitesimal imaginary parts chosen to select out the time-advanced or retarded Green function.

Since any practical calculation will involve some degree of approximation, it is important to ensure that this is consistent with that introduced for the one-particle Green function, in the sense that the resulting theory obeys the necessary conservation laws.

For the case of Schrödinger waves (e.g., electrons) prescriptions for $\langle\hat{\mathcal{G}}(z)\hat{\mathcal{G}}(z')\rangle$ consistent with the QCACP and the EMA have been developed by Roth and Singh [31] and by Itoh *et al.* [56–58,32–34]. Since classical waves exhibit a different energy conservation law, it is of interest to see to what extent these approaches may be applied to the present problem.

We choose to employ the "variational" method introduced by Roth and Singh [31] rather than the (equivalent) topological analysis of Itoh *et al.* [56]. In this, $\langle\hat{\mathcal{G}}(z)\hat{\mathcal{G}}(z')\rangle$ is identified with the variation $\delta\hat{G}(z,z')$ obtained via the following procedure.

Given a theory for the average amplitude Green function $\hat{G}(z)$, we make the variation $\hat{G}_0(z)\rightarrow\hat{G}_0(z)+\delta\hat{G}_0(z,z')$, where

$$\delta\hat{G}_0(z,z')=\hat{G}_0(z)\hat{G}_0(z'). \tag{44}$$

All quantities to the right of such variations are to be evaluated at frequency $z'$.

In this way, starting with Eq. (5), we find

$$\begin{aligned}\delta\hat{G}(z,z')&=\delta\hat{G}_0(z,z')+\delta\hat{G}_0(z,z')\hat{T}(z')\hat{G}_0(z')\\&\quad+G_0(z)\delta\hat{T}(z,z')\hat{G}_0(z')\\&\quad+\hat{G}_0(z)\hat{T}(z)\delta G_0(z,z')\\&=\hat{G}(z)\hat{G}(z')+\hat{G}_0(z)[\delta\hat{T}(z,z')\\&\quad-\hat{T}(z)\hat{G}_0(z)\hat{G}_0(z')\hat{T}(z')]\hat{G}_0(z').\end{aligned} \tag{45}$$

From the equation for the random $T$ matrix $\hat{\mathcal{T}}(z)$,

$$\hat{\mathcal{T}}(z)=\hat{V}(z)[1+\hat{G}_0(z)\hat{\mathcal{T}}(z)], \tag{46}$$

where the total potential $\hat{V}(z)=\sum_\alpha\hat{v}_\alpha(z)$, it is a simple matter to show that

$$\delta\hat{T}(z,z')=\langle\hat{\mathcal{T}}(z)\hat{G}_0(z)\hat{G}_0(z')\hat{\mathcal{T}}(z')\rangle, \tag{47}$$

so that, in the absence of correlations, $\delta\hat{G}$ reduces to the product of two average Green functions (as it should).

Energy conservation may be expressed by way of an identity connecting $\langle\hat{\mathcal{G}}(z)\hat{\mathcal{G}}(z')\rangle$ and the one-particle Green functions [59]. This is readily derived by considering the unaveraged Green function

$$\hat{\mathcal{G}}(z)=\hat{G}_0(z)+\hat{G}_0(z)\hat{V}(z)\hat{\mathcal{G}}(z). \tag{48}$$

Rewriting this in the form

$$[\hat{G}_0^{-1}(z)-\hat{V}(z)]\hat{\mathcal{G}}(z)=1 \tag{49}$$

and multiplying both sides by $\hat{V}(z')\hat{\mathcal{G}}(z')$, we arrive at

$$\hat{V}(z')\hat{\mathcal{G}}(z')[\hat{G}_0^{-1}(z)-\hat{V}(z)]\hat{\mathcal{G}}(z)=\hat{V}(z')\hat{\mathcal{G}}(z'). \tag{50}$$

Now, subtracting the equation obtained by interchanging $z$ and $z'$, it follows that

$$\begin{aligned}&\hat{V}(z')\hat{\mathcal{G}}(z')\hat{G}_0^{-1}(z)\hat{\mathcal{G}}(z)-\hat{V}(z)\hat{\mathcal{G}}(z)\hat{G}_0^{-1}(z')\hat{\mathcal{G}}(z')\\&\qquad=\hat{V}(z')\hat{\mathcal{G}}(z')-\hat{V}(z)\hat{\mathcal{G}}(z).\end{aligned} \tag{51}$$

In the case of energy-independent potentials the $\hat{V}$'s cancel and, after averaging, we find

$$\langle\hat{\mathcal{G}}(z)\hat{\mathcal{G}}(z')\rangle=[\hat{G}_0^{-1}(z')-\hat{G}_0^{-1}(z)]^{-1}[\hat{G}(z)-\hat{G}(z')]. \tag{52}$$

We may obtain a similar expression for potentials of the form

$$\hat{V}(z)=\phi(z)\hat{V}_0, \tag{53}$$

which includes those of interest here [see Eq. (36)], by defining the quantities

$$\hat{\tilde{\mathcal{G}}}(z) \equiv \phi(z)\hat{\mathcal{G}}(z),$$
$$\hat{\tilde{G}}_0(z) \equiv \phi(z)\hat{G}_0(z),$$
$$\hat{\tilde{G}}(z) \equiv \phi(z)\hat{G}(z), \tag{54}$$

so that

$$\langle\hat{\tilde{\mathcal{G}}}(z)\hat{\tilde{\mathcal{G}}}(z')\rangle = [\hat{\tilde{G}}_0^{-1}(z') - \hat{\tilde{G}}_0^{-1}(z)]^{-1}[\hat{\tilde{G}}(z) - \hat{\tilde{G}}(z')]. \tag{55}$$

It is helpful to rewrite this in terms of $\delta\hat{T}(z,z')$ and the average $T$ matrices. Using (5) it is a simple matter to deduce that

$$\delta\hat{T}(z,z') = [\hat{\tilde{G}}_0^{-1}(z') - \hat{\tilde{G}}_0^{-1}(z)]^{-1}[\hat{\tilde{T}}(z) - \hat{\tilde{T}}(z')], \tag{56}$$

where we define

$$\hat{\tilde{T}}(z) \equiv \hat{T}(z)/\phi(z). \tag{57}$$

The question is now whether $\delta\hat{T}(z,z')$, as derived via the procedure of Roth and Singh and Itoh *et al.*, satisfies (56).

Since $\hat{T} = \int\hat{\tau}(1,2)d(1)d(2)$, we may formulate this question in terms of $\delta\hat{\tau}(1,2)$. Recall that for an OZ-compatible theory this quantity satisfies an equation of the form (30), from which we may deduce

$$\delta\hat{\tau}(1,2) = \int\hat{\tau}(1,3)\hat{t}^{(m)-1}(3)\rho^{-1}\delta\hat{t}^{(m)}(3)\hat{t}^{(m)\prime-1}(3) \times\hat{\tau}'(3,2)d(3) + \int\hat{\tau}(1,3)\delta\hat{C}(3,4)\hat{\tau}'(4,2)d(3)d(4). \tag{58}$$

Here, for convenience, we have introduced a notation in which a prime denotes evaluation at complex frequency $z'$.

By writing $\hat{\tilde{\tau}}(1,2) \equiv \int\phi^{-1}\hat{\tau}(1,3)\delta(3,2)d(3)$, with $\delta(3,2)$ a Dirac delta function, and exploiting the fact that

$$\int[\delta(1,4)\rho^{-1}\hat{\tilde{t}}^{(m)-1}(1) - \hat{\tilde{C}}(1,4)]\hat{\tilde{\tau}}(4,3)d(4) = \delta(1,3), \tag{59}$$

where $\hat{\tilde{C}}(1,4) = \phi\hat{C}(1,4)$, we find for $\Delta\hat{\tilde{\tau}}(1,2) \equiv \hat{\tilde{\tau}}(1,2) - \hat{\tilde{\tau}}'(1,2)$,

$$\Delta\hat{\tilde{\tau}}(1,2) = \int\hat{\tilde{\tau}}(1,3)\hat{\tilde{t}}^{(m)-1}(3)\rho^{-1}\Delta\hat{\tilde{t}}^{(m)}(3) \times\hat{\tilde{t}}^{(m)\prime-1}(3)\hat{\tilde{\tau}}'(3,2)d(3) + \int\hat{\tilde{\tau}}(1,3)\Delta\hat{\tilde{C}}(3,4)\hat{\tilde{\tau}}'(4,2)d(3)d(4). \tag{60}$$

Comparing Eqs. (58) and (60), we see that the conservation law (56) will be satisfied as long as

$$\Delta\hat{\tilde{t}}^{(m)}(\alpha) = \hat{\tilde{G}}_0^{-1}\Delta\hat{\tilde{G}}_0\hat{\tilde{G}}_0^{\prime-1}\delta\hat{t}^{(m)}(\alpha), \tag{61a}$$
$$\Delta\hat{\tilde{C}}(\alpha,\beta) = \phi\phi'\hat{\tilde{G}}_0^{-1}\Delta\hat{\tilde{G}}_0\hat{\tilde{G}}_0^{\prime-1}\delta\hat{C}(\alpha,\beta). \tag{61b}$$

In view of the fact that, within the OZ formalism, a given theory is fully specified via the closure relation, we might expect to be able to express the conservation law as a single identity involving the direct propagator $\hat{\tilde{C}}(\alpha,\beta)$. For this, we need to demonstrate (61a) holds provided that (61b) is true.

Itoh *et al.* [32–34] define the quantity $\hat{\eta}(\alpha)$ (not to be confused with the packing fraction) in terms of which

$$\hat{t}^{(m)}(\alpha) = \hat{t}(\alpha)[1 + \hat{\eta}(\alpha)\hat{t}^{(m)}(\alpha)]. \tag{62}$$

From the discussion of Sec. III we see that, for an OZ-compatible theory

$$\hat{\eta}(\alpha) = \int\hat{H}(\alpha,\beta)\rho\hat{t}^{(m)}(\beta)\hat{G}_0d(\beta). \tag{63}$$

Using the fact that $\delta\hat{t}(\alpha) = \hat{t}(\alpha)\hat{G}_0\hat{G}_0'\hat{t}'(\alpha)$, we have

$$\delta\hat{t}^{(m)}(\alpha) = \hat{t}^{(m)}(\alpha)[\hat{G}_0\hat{G}_0' + \delta\hat{\eta}(\alpha)]\hat{t}^{(m)\prime}(\alpha). \tag{64}$$

Clearly now, satisfaction of (61a) is attained only if

$$\Delta\hat{\tilde{\eta}}(\alpha) \equiv \phi\hat{\eta}(\alpha) - \phi'\hat{\eta}'(\alpha) = \phi\phi'\hat{\tilde{G}}_0^{-1}\Delta\hat{\tilde{G}}_0\hat{\tilde{G}}_0^{\prime-1}\delta\hat{\eta}(\alpha). \tag{65}$$

From (63) it follows that this relies on the identity [with $\Delta\hat{\tilde{H}}(1,2)$ similarly defined]

$$\Delta\hat{\tilde{H}}(1,2) = \phi\phi'\hat{\tilde{G}}_0^{-1}\Delta\hat{\tilde{G}}_0\hat{\tilde{G}}_0^{\prime-1}\delta\hat{H}(1,2). \tag{66}$$

Analogous arguments, based on the OZ relation (19), may be used to establish that this requires satisfaction of both (61a) and (61b). However, we have already determined that (61a) relies on (66). By a process of iteration it follows, from a term-by-term comparison, that, as expected, we need only demonstrate that (61b) holds.

For a given theory this is determined by examining the closure relation, which for the EMA leads to

$$\delta\hat{C}(1,2) = g_2(1,2)\delta\hat{G}_0 + \rho h_2(1,2)\Big\{\int\delta\hat{C}(1,3) \times\hat{t}^{(m)\prime}(3)\hat{H}'(3,2)d(3) + \int\hat{C}(1,3)\delta\hat{t}^{(m)}(3)\hat{H}'(3,2)d(3) + \int\hat{C}(1,3)\hat{t}^{(m)\prime}(3)\delta\hat{H}'(3,2)d(3)\Big\}, \tag{67}$$

$$\Delta\hat{\tilde{C}}(1,2) = g_2(1,2)\Delta\hat{\tilde{G}}_0 + \rho h_2(1,2)\Big\{\int\Delta\hat{\tilde{C}}(1,3) \times\hat{\tilde{t}}^{(m)\prime}(3)\hat{\tilde{H}}'(3,2)d(3) + \int\hat{\tilde{C}}(1,3)\Delta\hat{\tilde{t}}^{(m)}(3)\hat{\tilde{H}}'(3,2)d(3) + \int\hat{\tilde{C}}(1,3)\hat{\tilde{t}}^{(m)}(3)\Delta\hat{\tilde{H}}(3,2)d(3)\Big\}. \tag{68}$$

Expanding all occurrences of $\Delta\hat{\tilde{H}}$, $\delta\hat{H}$, $\Delta\hat{\tilde{t}}^{(m)}$, and $\delta\hat{t}^{(m)}$ in terms of $\Delta\hat{\tilde{C}}$ and $\delta\hat{C}$ and iterating the resulting equations readily confirms that Eq. (61b) holds.

The same may also be verified for the correlated Elyutin theory. As mentioned in Sec. III, this theory may be of particular relevance for studying localization. This may be ascertained from a diagrammatic analysis (cf. Itoh and Watabe [57]), which reveals that it incorporates maximally crossed diagrams, known to be important in that context.

For the EMA Itoh *et al.* [32–34] have considered in some detail how the above formalism may be employed in calculating the electronic conductivity. In particular, they have shown how the off-shell parts may be partially decoupled from the rest of the problem. For wave transport the quantity of interest is $\Phi_{\mathbf{p},\mathbf{p}'}(\mathbf{q},\omega,\Delta\omega) \equiv \langle G(\mathbf{p}+\mathbf{q}/2,\omega+\mathrm{i}0+\Delta\omega/2)G(\mathbf{p}'-\mathbf{q}/2,\omega-\mathrm{i}0-\Delta\omega/2)\rangle$. By examining its asymptotic behavior in the limit $\Delta\omega, q \to 0$, we may extract a diffusion constant [59]. The details of how this may be achieved within the present formalism will be considered elsewhere.

## V. CONCLUSIONS

In this paper we have written down a general formalism within which one may develop theories for both the amplitude and intensity Green functions in such a way as to guarantee energy conservation. Previous work in this area has often been achieved by neglecting positional correlations among the scatterers [9,10,12] or by approximating them in a rather *ad hoc* fashion [60–63]. The advantage of the present formalism is that it allows the effects of multiple scattering and of scatterer correlations to be incorporated on an equal footing.

Within this framework the EMA is identified as the simplest in a class of theories for the amplitude Green function $\hat{G}$ that preserve the physical requirement that the scatterers should not overlap. This enables a decoupling of the on- and off-shell parts of $\hat{G}$. Higher-order approximations within this class permit the systematic inclusion of recurrent scattering terms. Similar terms have been introduced in the context of point scatterer models in an effort to study the effects of dependent scattering. Where the scattering cross section $\sigma_s$ of the individual scatterers greatly exceeds their geometrical cross section $\pi a^2$, the regime of physical interest is shifted to a sufficiently low packing fraction $\eta$ that the neglect of positional correlations is likely to be justifiable. However, given typical values of $\sigma_s \approx 6\pi a^2$, for dielectric scatterers, such correlations are likely to be important if one is to make quantitative comparisons with experiment. The present framework enables one to incorporate these and the recurrent scattering terms on an equal basis. An investigation of their relative importance will be given in a future paper.

In addition, as will be shown in a future paper, the analogy with liquid-state theory may be exploited further for isotropic scatterers to obtain analytical results for a simple (step-function) model pair correlation function $g_2(\alpha,\beta)$. As will also be demonstrated, the methods involved in deriving such a solution suggest slightly modified closure approximations that may be used in numerical work when more realistic forms for $g_2(\alpha,\beta)$ are adopted.

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