Generalization of the Foldy-Lax Formula for the Self-Energy of a Wave Propagating in a Disordered System of Scatterers

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We present an exact generalization of the Foldy-Lax formula for the self-energy of a wave propagating in a disordered system of identical spherical scatterers. The Foldy-Lax formula yields an expression for the self-energy valid to first order in the density of scatterers. Our exact formula allows a systematic calculation of corrections to this low-density approximation. The formula is based on a renormalized cluster expansion which was presented earlier.

KEY WORDS: Multiple scattering; disordered systems; Foldy-Lax formula; cluster expansion.

1. INTRODUCTION

The simplest approximation to the optical potential or self-energy of a wave propagating in a disordered system of spherical scatterers is given by the Foldy–Lax formula.⁽¹⁻³⁾ One obtains this formula by adding the forward scattering amplitudes from the individual scatterers, assumed distributed randomly, and neglecting multiple scattering. The approximation is valid to first order in the density n of scatterers. Explicitly the Foldy–Lax formula for the wave-vector- and energy-dependent self-energy reads

$$\Sigma(\mathbf{q}, E) = n(\mathbf{q} | M(E) | \mathbf{q}) \tag{1.1}$$

where M(E) is the one-body T-matrix of a single scatterer taken to be centered at the origin. Here we have employed the notation

$$(\mathbf{q}|A|\mathbf{q}') = \int \exp(-i\mathbf{q}\cdot\mathbf{r}) A(\mathbf{r},\mathbf{r}') \exp(i\mathbf{q}'\cdot\mathbf{r}') d\mathbf{r} d\mathbf{r}'$$
(1.2)

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Since we need the forward scattering amplitude, both wave vectors in (1.1) are equal.

In this paper we present a generalization of the Foldy–Lax formula which in principle is exact and valid to arbitrary order in the density. Our formula is based on the renormalized cluster expansion which we have presented earlier.⁽⁴⁾ We assume that the disordered system of scatterers on average is spatially uniform in the thermodynamic limit. Exploiting this assumption and using Fourier transformation, we are led to the concise formula

$$\Sigma(\mathbf{q}, E) = n(\mathbf{q} | M(E) [I - nR(\mathbf{q}, E) M(E)]^{-1} | \mathbf{q})$$
(1.3)

where $R(\mathbf{q}, E)$ is a linear operator which we call the recurrence operator. Although this operator cannot be evaluated explicitly for a dense system of scatterers, it can be expressed exactly in terms of a cluster expansion. Thus, (1.3) is a useful starting point for the calculation of corrections to the Foldy-Lax formula.

In Section 5 we present a generalization of the self-energy which we have found useful in work in linear hydrodynamics, where similar concepts apply. For this generalized self-energy we derive an expression analogous to (1.3).

The general theory has numerous applications.⁽⁵⁻⁷⁾ We have already studied the effective dielectric constant of nonpolar fluids and suspensions.⁽⁸⁾ Other applications of the theory will be published elsewhere.

2. SUMMARY OF BASIC EQUATIONS

We consider an *m*-component vector wave propagating in a disordered static array of identical scatterers. The time-independent linear wave equation for a particular configuration of scatterers reads

$$\mathscr{L}_{0}\psi(1,...,N;\mathbf{r}) - V(1,...,N)\psi(1,...,N;\mathbf{r}) = s(\mathbf{r})$$
(2.1)

where \mathcal{L}_0 is the wave operator for the uniform medium, and V(1,..., N) is the potential operator, which depends on the configuration of scatterers. We assume that the scattering potential consists of a sum of identical onebody terms

$$V(1,...,N) = \sum_{j=1}^{N} V(j)$$
(2.2)

The label j in V(j) indicates the parametric dependence on the position of the scattering center \mathbf{R}_j and possibly orientational variables Ω_j .

We assume that the disorder of the system is described by a known probability distribution W(1,..., N). Moreover, we assume that the system in volume Ω on average is spatially uniform and possesses a well-defined thermodynamic limit $N \to \infty$, $\Omega \to \infty$ with uniform density n(1) and translationally invariant partial distribution functions n(1,..., s).

Let

$$\psi_0(\mathbf{r}) = \int G_0(\mathbf{r} - \mathbf{r}') \cdot s(\mathbf{r}') \, d\mathbf{r}' \tag{2.3}$$

be the solution in the absence of scatterers, where $G_0(\mathbf{r} - \mathbf{r}')$ is the freespace Green function corresponding to outgoing waves. Defining the *N*-body *T*-matrix by

$$V(1,...,N)\psi(1,...,N) = T(1,...,N)\psi_0$$
(2.4)

we obtain for the self-energy operator X of the average wave, defined by $\langle V\psi \rangle = X \langle \psi \rangle$,

$$X = \langle T \rangle (I + G_0 \langle T \rangle)^{-1} \tag{2.5}$$

where the average is over the probability distribution W(1,...,N).

Due to (2.2) the *N*-body *T*-matrix has the multiple scattering expansion

$$T(1,...,N) = \sum_{j=1}^{N} M(j) + \sum_{l=2}^{\infty} \sum_{[j]}^{\prime} M(j_{1}) \prod_{l=2}^{l} G_{0}M(j_{l})$$
(2.6)

where M(j) is the one-body T-matrix for scatterer j and the last sum is over all sequences [j] of l labels with the condition, indicated by the prime, that no label be repeated in succession. We have shown in I on the basis of the multiple scattering expansion that the average T-matrix may be cast in the form

$$\langle T \rangle = \int d1 \, n(1) \, B(1) + \int d1 \, d2 \, n(1) \, n(2) \, B(1) \, H(1, 2) \, B(2)$$
 (2.7)

where the bridge operator B(1) is given by

$$B(1) = M(1)[I - S(1) M(1)]^{-1}$$
(2.8)

with the reaction field operator S(1). The pair connector H(1, 2) is expressed by an Ornstein-Zernike type equation

$$H(1,2) = C(1,2) + \int d3 \, n(3) \, C(1,3) \, B(3) \, H(3,2) \tag{2.9}$$

in terms of the direct connector C(1, 2). The latter is related to the short-range connector S(1, 2) by

$$C(1, 2) = G_0 + S(1, 2) \tag{2.10}$$

The reaction field operator S(1) and the short-range connector S(1, 2) are the basic constituents of the theory. Each of these operators may be decomposed in terms of a cluster expansion

$$S(1) = \sum_{s=2}^{\infty} S_s(1), \qquad S(1,2) = \sum_{s=2}^{\infty} S_s(1,2)$$
(2.11)

where $S_s(1)$ and $S_s(1, 2)$ are given as integrals of s-body scattering operators over s-body correlation functions. The detailed expressions for these cluster operators have been given in I. As an example, we repeat here the expressions for the two-body terms $S_2(1)$ and $S_2(1, 2)$. The two-body contribution $S_2(1)$ is given by

$$S_2(1) = \int d2 \, n(2) \, g(1, 2) \, N_{11}(1, 2) \tag{2.12}$$

where g(1, 2) is the pair distribution function and $N_{11}(1, 2)$ is a nodal connector which may be found from the solution of the two-body scattering problem. The connector $N_{11}(1, 2)$ summarizes all scattering processes between the particles 1 and 2 for which the first and the last scatterer are 1. Similarly, the two-body contribution $S_2(1, 2)$ is given by

$$S_2(1, 2) = g(1, 2)[N_{12}(1, 2) - G_0] + h(1, 2) G_0$$
(2.13)

where $N_{12}(1, 2)$ is a second nodal connector and h(1, 2) = g(1, 2) - 1 is the pair correlation function. The connector $N_{12}(1, 2)$ summarizes all scattering processes between the particles 1 and 2 for which the first scatterer is 2 and the last scatterer is 1. The higher order terms in (2.11) are defined in similar manner. As the order increases, the terms become more and more difficult to evaluate. In the present paper we are not concerned with the explicit calculation, but derive a general and exact result for the self-energy, valid for spherical particles.

We have shown in I that the self-energy operator X, given by (2.5), may be expressed as

$$X = \int d1 \ n(1) \ B(1) + \int d1 \ d2 \ n(1) \ n(2) \ B(1) \ F(1, 2) \ B(2)$$
 (2.14)

where the pair connector F(1, 2) is related to the short-range connector S(1, 2) by the Ornstein-Zernike type equation

$$F(1, 2) = S(1, 2) + \int d3 \, n(3) \, S(1, 3) \, B(3) \, F(3, 2) \tag{2.15}$$

In principle, a calculation of the operators S(1) and S(1, 2) yields the selfenergy operator X via (2.8), (2.14), and (2.15).

3. SUSCEPTIBILITY OPERATOR AND SELF-ENERGY

The relations given above are valid for a finite system of N scatterers enclosed in a finite volume Ω . They remain valid when boundary conditions on the wavefunction are imposed at the walls of Ω , provided the onebody T-matrix M and the Green function G_0 are changed accordingly. In practice one is usually interested in results valid in the thermodynamic limit. Averaging the wave equation (2.1), one obtains for the average wave

$$\mathscr{L}_0\langle\psi\rangle - X\langle\psi\rangle = s \tag{3.1}$$

We expect that in the thermodynamic limit for a spatially uniform system the self-energy operator X becomes translationally invariant with a kernel $X_{\infty}(\mathbf{r} - \mathbf{r}')$ which depends only on the difference $\mathbf{r} - \mathbf{r}'$. On the other hand, the solution of (3.1)

$$\langle \psi(\mathbf{r}) \rangle = \int G_{\mathrm{av}}(\mathbf{r}, \mathbf{r}') \cdot s(\mathbf{r}') \, d\mathbf{r}'$$
 (3.2)

in general will not possess a thermodynamic limit independent of the shape of Ω . Nonetheless, for fixed arguments **r** and **r'** the average Green function $G_{av}(\mathbf{r}, \mathbf{r'})$ will tend to a limit $G_{av}^{\infty}(\mathbf{r} - \mathbf{r'})$ depending only on the difference $\mathbf{r} - \mathbf{r'}$. Formally, G_{av}^{∞} is simply the inverse $[\mathscr{L}_0 - X_{\infty}]^{-1}$. In general this operator will have a long-range dependence on the distance $|\mathbf{r} - \mathbf{r'}|$.

It is evident from (2.14) that the self-energy operator X may alternatively be written in the form

$$X = \int d1 \ n(1) \ \chi(1)$$
 (3.3)

with the susceptibility operator $\chi(1)$ given by

$$\chi(1) = B(1) + \int d2 \, n(2) \, B(1) \, F(1, 2) \, B(2) \tag{3.4}$$

We may expect that the corresponding kernel $\chi(1; \mathbf{r}, \mathbf{r}')$ is of short range in the variables $\mathbf{r} - \mathbf{R}_1$ and $\mathbf{r}' - \mathbf{R}_1$, so that the action of the operator $\chi(1)$ is localized about the position \mathbf{R}_1 . On the other hand, if we write the average *T*-matrix in the same form

$$\langle T \rangle = \int d1 \ n(1) \ \tau(1)$$
 (3.5)

with, according to (2.7),

$$\tau(1) = B(1) + \int d2 \, n(2) \, B(1) \, H(1, 2) \, B(2) \tag{3.6}$$

then the kernel $\tau(1; \mathbf{r}, \mathbf{r}')$ is of long range. Using the equations of the preceding section and the Dyson equation

$$G_{\rm av} = G_0 + G_0 X G_{\rm av} \tag{3.7}$$

one finds that the operators $\tau(1)$ and $\chi(1)$ are related by

$$\tau(1) = \chi(1) + \int d2 \, n(2) \, \chi(1) \, G_{\rm av} \, \chi(2) \tag{3.8}$$

Therefore the long-range properties of the operator $\tau(1)$ are directly related to those of the average Green function G_{av} .

The self-energy $\chi_{\infty}(\mathbf{q})$ is defined in the thermodynamic limit by

$$\lim_{\substack{N \to \infty \\ a \to \infty}} (\mathbf{q} | X | \mathbf{q}') = 8\pi^3 \chi_{\infty}(\mathbf{q}) \,\delta(\mathbf{q} - \mathbf{q}')$$
(3.9)

In (1.1) and (1.3) we have employed a more conventional notation for the self-energy. Note that in general $\chi_{\infty}(\mathbf{q})$ is a tensor of rank *m*. The delta function in (3.9) is an expression of the translational invariance of the system in the thermodynamic limit.

4. RECURRENCE OPERATOR

In order to simplify the discussion, we assume henceforth that the particles are spherical. Thus, in order to specify a particular condiguration of scatterers, it suffices to specify the positions $(\mathbf{R}_1,...,\mathbf{R}_N)$ of the sphere centers. The integration symbol dj may be replaced by $d\mathbf{R}_j$. In the thermodynamic limit the susceptibility operator $\chi(1)$, defined in (3.4), will be translationally invariant and the corresponding kernel will have the form

$$\lim_{\substack{N \to \infty \\ \Omega \to \infty}} \chi(1) = \chi(\mathbf{r} - \mathbf{R}_1, \mathbf{r}' - \mathbf{R}_1)$$
(4.1)

From (3.3) and the definition (3.9) we find

$$\chi_{\infty}(\mathbf{q}) = \lim_{\substack{N \to \infty \\ \Omega \to \infty}} n(1)(\mathbf{q} | \chi(1) | \mathbf{q}) |_{\mathbf{R}_1 = 0}$$
(4.2)

We evaluate the right-hand side by substituting (3.4). Here we use the fact that in the thermodynamic limit the pair connector F(1, 2) depends on its variables \mathbf{R}_1 , \mathbf{R}_2 , \mathbf{r} , \mathbf{r}' in the translationally invariant way

$$\lim_{\substack{N \to \infty \\ \Omega \to \infty}} F(1, 2) = F(\mathbf{R}_2 - \mathbf{R}_1; \mathbf{r} - \mathbf{R}_1, \mathbf{r}' - \mathbf{R}_2)$$
(4.3)

As in (4.1), we do not indicate the thermodynamic limit explicitly on the right-hand side. In the same limit the operators B(1) and B(2) in (3.4) have a dependence like (4.1). Employing variables of integration defined relative to the two centers, we find

$$\chi_{\infty}(\mathbf{q}) = n(\mathbf{q} |B| \mathbf{q}) + n^{2}(\mathbf{q} |BF(\mathbf{q}) B| \mathbf{q})$$
(4.4)

where the operators B are centered at the origin and $F(\mathbf{q})$ is the Fourier-transformed kernel

$$F(\mathbf{q};\mathbf{s},\mathbf{s}') = \int d\mathbf{R}[\exp(i\mathbf{q}\cdot\mathbf{R})] F(\mathbf{R};\mathbf{s},\mathbf{s}')$$
(4.5)

with $\mathbf{R} = \mathbf{R}_2 - \mathbf{R}_1$, $\mathbf{s} = \mathbf{r} - \mathbf{R}_1$, and $\mathbf{s}' = \mathbf{r}' - \mathbf{R}_2$. Fourier transforming the integral equation (2.15) and taking the thermodynamic limit, we find

$$F(\mathbf{q}) = S^{(2)}(\mathbf{q}) + nS^{(2)}(\mathbf{q}) BF(\mathbf{q})$$
(4.6)

where the operator $S^{(2)}(\mathbf{q})$ is defined as in (4.5) from the kernel S(1, 2). The superscript serves as a reminder of the distinction between the operators S(1, 2) and S(1). Equation (4.6) has the formal solution

$$F(\mathbf{q}) = [I - nS^{(2)}(\mathbf{q}) B]^{-1} S^{(2)}(\mathbf{q})$$
(4.7)

Substituting this in (4.4), we obtain

$$\chi_{\infty}(\mathbf{q}) = n(\mathbf{q} | B(I - nS^{(2)}(\mathbf{q}) | B)^{-1} | \mathbf{q})$$
(4.8)

Finally, we find upon substitution of (2.8)

$$\chi_{\infty}(\mathbf{q}) = n(\mathbf{q} | M(I - nR(\mathbf{q}) | M)^{-1} | \mathbf{q})$$
(4.9)

with the operator $R(\mathbf{q})$ given by

$$nR(\mathbf{q}) = S^{(1)} + nS^{(2)}(\mathbf{q}) \tag{4.10}$$

where $S^{(1)}$ is defined by

$$S^{(1)} = \lim_{\substack{N \to \infty \\ \Omega \to \infty}} S(1)|_{\mathbf{R}_1 = 0}$$
(4.11)

It may be worthwhile to write the important relation (4.10) more explicitly as a relation between integral kernels

$$nR(\mathbf{q}; \mathbf{s}, \mathbf{s}') = S^{(1)}(\mathbf{s}, \mathbf{s}') + nS^{(2)}(\mathbf{q}; \mathbf{s}, \mathbf{s}')$$
(4.12)

where in the second term on the right the field variables s and s' have been defined following (4.5).

The expression (4.9) is an important key to further calculations. It shows explicitly how the elementary constituent operators $S^{(1)}$ and $S^{(2)}(\mathbf{q})$ act in recurrent fashion. For this reason we call $R(\mathbf{q})$ the recurrence operator.

5. GENERALIZED SUSCEPTIBILITY

In work in linear hydrodynamics, where the same concepts apply, we have found it useful to extend the formalism and to introduce more general susceptibilities. We begin by noting that the self-energy may be regarded alternatively as a susceptibility. Introducing the Fourier transforms

$$\langle \psi \rangle_{\mathbf{q}} = \frac{1}{8\pi^3} \int \langle \psi \rangle(\mathbf{r}) \exp(-i\mathbf{q} \cdot \mathbf{r}) d\mathbf{r}$$
$$\langle V\psi \rangle_{\mathbf{q}} = \frac{1}{8\pi^3} \int \langle V\psi \rangle(\mathbf{r}) \exp(-i\mathbf{q} \cdot \mathbf{r}) d\mathbf{r}$$
(5.1)

we find from the definition (3.9) in the thermodynamic limit

$$\langle V\psi \rangle_{\mathbf{q}} = \chi_{\infty}(\mathbf{q}) \langle \psi \rangle_{\mathbf{q}} \tag{5.2}$$

We recall that $\chi_{\infty}(\mathbf{q})$ may be found from (3.4) and (4.2). The response to the applied field ψ_0 is given more directly by

$$\langle V\psi\rangle = \langle T\rangle\psi_0 \tag{5.3}$$

but this relation is highly nonlocal.

We generalize the susceptibility concept by introducing a modified T-operator via the multiple scattering expansion

$$T_{JK}(1,...,N) = \sum_{j=1}^{N} M_{JK}(j) + \sum_{\substack{j=1\\k \neq j}}^{N} J(j) G_0 K(k) + \sum_{l=2}^{\infty} \sum_{[j]}^{\prime} J(j_1) \left[\prod_{i=2}^{l} G_0 M(j_i) \right] G_0 K(j_{l+1})$$
(5.4)

where M_{JK} , J, and K are three specified one-body operators and the last sum is over all sequences [j] of l+1 labels with the condition that no label be repeated in succession. When M_{JK} , J, and K are all identical with the M operator, then T_{JK} reduces to the T-matrix as given by (2.6). On the basis of the above multiple scattering expansion we may derive results analogous to those of Section 2.

In particular, we find that (2.7) is generalized to

$$\langle T_{JK} \rangle = \int d1 \, n(1) \, B_{JK}(1) + \int d1 \, d2 \, n(1) \, n(2) \, B_J(1) \, H(1, 2) \, B_K(2)$$
 (5.5)

with the first bridge operator defined by

$$B_{JK}(1) = M_{JK}(1) + B_J(1) S(1) K(1) = M_{JK}(1) + J(1) S(1) B_K(1)$$
(5.6)

and the other two bridge operators defined by

$$B_{J}(1) = J(1)[I - S(1) M(1)]^{-1}$$

$$B_{K}(1) = [I - M(1) S(1)]^{-1} K(1)$$
(5.7)

Corresponding to (5.5) we define

$$X_{JK} = \int d1 \ n(1) \ \chi_{JK}(1) \tag{5.8}$$

with the susceptibility operator

$$\chi_{JK}(1) = B_{JK}(1) + \int d2 \, n(2) \, B_J(1) \, F(1, 2) \, B_K(2) \tag{5.9}$$

In analogy to (3.9) and (4.2), we obtain in the thermodynamic limit

$$\lim_{\substack{N \to \infty \\ \mathbf{q} \to \infty}} (\mathbf{q} | X_{JK} | \mathbf{q}') = 8\pi^3 \chi_{JK}^{\infty}(\mathbf{q}) \,\delta(\mathbf{q} - \mathbf{q}')$$
(5.10)

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with the generalized susceptibility

$$\chi_{JK}^{\infty}(\mathbf{q}) = \lim_{\substack{N \to \infty \\ \Omega \to \infty}} n(1)(\mathbf{q} | \chi_{JK}(1) | \mathbf{q}) |_{\mathbf{R}_1 = 0}$$
(5.11)

As the generalization of (4.4) we find

$$\chi_{JK}^{\infty}(\mathbf{q}) = n(\mathbf{q} | B_{JK} | \mathbf{q}) + n^2(\mathbf{q} | B_J F(\mathbf{q}) | B_K | \mathbf{q})$$
(5.12)

where the operators B_{JK} , B_J , and B_K are centered at the origin. By use of some operator algebra and the relations (4.7), (4.10), (5.6), and (5.7) this may be cast in the form

$$\chi_{JK}^{\infty}(\mathbf{q}) = n(\mathbf{q} \mid M_{JK} \mid \mathbf{q}) + n^{2}(\mathbf{q} \mid JR(\mathbf{q})[I - nMR(\mathbf{q})]^{-1} K \mid \mathbf{q}) \quad (5.13)$$

It is easily checked that this reduces to (4.9) when M_{JK} , J, and K all equal M.

We note that in analogy to (3.5) we may write

$$\langle T_{JK} \rangle = \int d1 \, n(1) \, \tau_{JK}(1) \tag{5.14}$$

with the operator

$$\tau_{JK}(1) = B_{JK}(1) + \int d2 \, n(2) \, B_J(1) \, H(1, 2) \, B_K(2) \tag{5.15}$$

By the same derivation which leads to (3.8), it follows that

$$\tau_{JK}(1) = \chi_{JK}(1) + \int d2 \ n(2) \ \chi_{JM}(1) \ G_{av} \chi_{MK}(2)$$
(5.16)

where $\chi_{JM}(1)$ and $\chi_{MK}(1)$ are defined by (5.9) with $M_{JM} = J$ and $M_{MK} = K$. This shows that the response kernel $\tau_{JK}(1)$ has long-range properties caused by the presence of the average Green function G_{av} . In addition, (5.16) provides an efficient way of calculating the response operator $\tau_{JK}(1)$. The susceptibility operators appearing in (5.16) have short-range kernels which may be evaluated in the thermodynamic limit from (5.13). In a separate paper we shall demonstrate the use of (5.16) in a calculation of the collective diffusivity of a system of Brownian particles with hydrodynamic interactions.

6. DISCUSSION

We have derived an exact generalization of the Foldy-Lax formula for the self-energy of a wave propagating in a disordered system of identical

spherical scatterers. Our exact formula is given in Eq. (4.9). The recurrence operator $R(\mathbf{q})$ appearing in this equation may in principle be calculated from a cluster expansion. In a preceding paper⁽⁴⁾ we have presented a detailed derivation of the cluster expansion. It is also easy to formulate several familiar approximation schemes within the present framework. For example, the ring approximation of Foldy and Walecka⁽⁹⁾ amounts to approximating the recurrence operator by

$$nR(\mathbf{q}) \approx G_n - G_0 \tag{6.1}$$

independent of \mathbf{q} , where G_n is the renormalized Green function found from the approximate Dyson equation

$$G_n = G_0 + G_0 n \overline{M} G_n \tag{6.2}$$

with the integrated one-body T-matrix

.

$$\bar{M} = \int d\mathbf{R}_1 \ M(1) \tag{6.3}$$

Another well-known approximation is the quasicrystalline approximation of Lax.^(2,3) In this approximation the recurrence operator is approximated by

$$R(\mathbf{q};\mathbf{s},\mathbf{s}') \approx \int h(\mathbf{R}) G_0(\mathbf{s}-\mathbf{s}'-\mathbf{R}) \exp(i\mathbf{q}\cdot\mathbf{R}) d\mathbf{R}$$
(6.4)

where $h(\mathbf{R})$ is the pair correlation function. It is clear that both approximations (6.1) and (6.4) are rather crude and that a more sophisticated approach is needed. Self-consistent schemes, such as the coherent potential approximation,^(10,11) are not easily explained within the present framework. To that purpose it is necessary to first formulate the exact theory in a selfconsistent fashion.

In Section 5 we have extended the theory in a manner which we have found useful in problems in linear hydrodynamics, such as the sedimentation of spheres in a quiescent fluid and the effective viscosity of suspensions of spheres. These applications of the general theory, as well as a comparison with results from various approximation schemes, will be presented elsewhere. $^{(12, 13)}$

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