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Microscopic Approach to the Lorentz Cavity in Dielectrics

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We develop a microscopic scattering theory for the electromagnetic response of dielectrics. We derive the Lorentz-Lorenz relation for a hard-sphere fluid and for hard-sphere mixtures by summing rigorously the relevant class of multiple scattering events which incorporates particle correlations. The derivation neither makes use of macroscopic concepts such as local and reaction fields nor does it invoke decoupling schemes for high-order correlation functions. [S0031-9007(97)03660-0]

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Understanding the relation between macroscopic electromagnetic response as observed in experiments and microscopic properties such as electronic, atomic, and molecular polarizabilities still presents a challenge. This fundamental problem is relevant at any frequency of the electromagnetic field—from electrostatics up to the x-ray region. Its solution is fierce as it involves a genuine many-body problem. Different forms of matter—metals, semiconductors, insulators, liquids, plasmas, gases, and mesoscopic structures—and different ranges of frequencies are expected to require their own specific solution. However, in 1880 Lorentz [1] developed a generic theory of superior quality.

Within the framework of standard response theory, the dielectric constant ϵ is given by

$$\epsilon = 1 + \rho\alpha, \quad (1)$$

in which α is the polarizability and ρ the density of the microscopic constituents. (As the polarizability α depends on the angular frequency ω of the electric field, ϵ will also depend on ω . The obvious dependence on ω in all equations will be dropped. We use rationalized Gaussian units and $c_0 = 1$.) Lorentz demonstrated that a dramatic improvement over Eq. (1) can be obtained at all frequencies by *postulating* the existence of a dynamic “local field.” This local field differs from the macroscopic field by a correction factor: the Lorentz local-field factor. In its simplest form this factor is given by $(\epsilon + 2)/3$. Applying this local-field correction to the dielectric constant results in

$$\epsilon = 1 + \rho\alpha \frac{\epsilon + 2}{3}, \quad (2)$$

which is the Lorentz-Lorenz relation (LLR). Historically, the LLR refers to formula (2) when solved explicitly for the polarizability α . Its zero-frequency version is often called the Clausius-Mossotti equation [2]. The success of the LLR in predicting the dielectric constant, in many cases within 1% accuracy, is impressive [3].

From the point of view of many-body physics, the LLR is quite surprising as can best be appreciated by solving

Eq. (2) for ϵ ,

$$\epsilon = 1 + \rho\alpha \frac{1}{1 - \frac{1}{3}\rho\alpha}. \quad (3)$$

Apparently, the local-field concept involves terms up to infinite order in the density. The LLR is not expected to be exact, and one would like to know which many-body contributions are included and which are not. To get more insight into the range of validity of the LLR, many alternative or simplified derivations of the original result by Lorentz have been presented over the past century. We mention only a few: Born and Wolf [2], Debye [4], Onsager [5], De Goede and Mazur [6]. Van Kranendonk and Sipe [7] and Schnatterly and Tarrío [8] have reviewed a large number of these derivations. Unfortunately, these approaches rely on the concept of a local field and then apply macroscopic arguments based on Maxwell’s equations. The theoretical base of the LLR is therefore hard to assess, and thus difficult to improve on. We know of no exact microscopic derivation of the LLR, and this Letter is aimed at filling this gap. Microscopic derivations of the second-order term $\frac{1}{3}\rho^2\alpha^2$ of (3) employing point dipoles have already been obtained by Kuz’min *et al.* [9] and Morice *et al.* [10].

The electric field seen by one particular point dipole i can be written schematically as [2]

$$\mathbf{E}_i = \mathbf{E}_{\text{inc}} + \sum_{j \neq i} \mathbf{E}_{ij}, \quad (4)$$

where \mathbf{E}_{inc} is the incoming field and \mathbf{E}_{ij} is the field received by dipole i radiated from dipole j . Equation (4) in itself is exact, but the summation over dipoles j is very difficult to carry out. In the conventional derivations (see, e.g., [2]) it is assumed that the positions of the j dipoles can be replaced by a continuous dipole density. With this assumption the summation turns into an integration,

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_{\text{inc}}(\mathbf{r}) + \int' d\mathbf{r}_1 \mathbf{E}(\mathbf{r}, \mathbf{r}_1). \quad (5)$$

To include some remnant of the particle correlations this integration is performed by excluding a small sphere—the Lorentz cavity—around the origin (indicated by the prime). The electric fields in Eq. (5) induce polarizations that can be represented by the dielectric dyadic ϵ ,

$$\epsilon - \mathbf{I} = \rho \alpha \mathbf{I} - \rho \alpha \omega^2 \int' d\mathbf{r} \mathbf{G}_0(\mathbf{r}) \cdot [\epsilon - \mathbf{I}], \quad (6)$$

in which \mathbf{I} is the unit dyadic and \mathbf{G}_0 the free-space Green's function, to be specified below, signifying the propagation of the scattered light waves. This self-consistent equation for ϵ can be solved and leads to the LLR. This type of intuitive derivation has *hitherto* been the sole theoretical foundation for the LLR. One of the shortcomings of these derivations of the LLR is readily identified: All light scattering in which more than two correlated particles are involved has been replaced by contributions in which only products of pair correlations occur.

The following derivation of the LLR takes into account *all* particle correlations rigorously and, indeed, up to infinite order. We shall also be able to generalize the microscopic theory for the dielectric response to mixtures of hard spheres, and we will derive the LLR for mixtures [3], which is closely related to the Maxwell-Garnett formula [11].

The basis of our approach is multiple-scattering theory. The generic microscopic building block that we consider is the finite-size hard sphere with radius R (being much smaller than the wavelength of light) and polarizability α [12]. Our analysis includes the case of the point dipole [13].

The dielectric function originates from the collective response of dipoles excited by the incoming field and the rescattered fields of all other dipoles. The scattering of the three electric field components from position \mathbf{r}_1 to position \mathbf{r}_2 by a particle located at \mathbf{R}_i is completely described by the t matrix, denoted by the second-rank tensor $\mathbf{t}_i(\mathbf{r}_1, \mathbf{r}_2)$. For a spherical Rayleigh scatterer this t matrix is given by

$$\mathbf{t}_i(\mathbf{r}_1, \mathbf{r}_2) = -\frac{\alpha \omega^2}{v} \delta(\mathbf{r}_1 - \mathbf{r}_2) \theta(R - |\mathbf{r}_1 - \mathbf{R}_i|) \mathbf{I}, \quad (7)$$

in which θ denotes the Heaviside step function and $v = 4\pi R^3/3$ is the volume of the scatterer. The step function ensures that the scattering takes place only when the light has propagated into the scatterer, and the delta function signifies that in the Rayleigh limit, as in the Born approximation, the wave only interacts once with the scattering potential. From now on, $\hat{\theta}_i(\mathbf{r}) \equiv \theta(R - |\mathbf{r} - \mathbf{R}_i|)$. The free-field propagator $\mathbf{G}_0(\mathbf{r}_1, \mathbf{r}_2) = \mathbf{G}_0(\mathbf{r}_1 - \mathbf{r}_2)$ describes how the electric field propagates from \mathbf{r}_1 to \mathbf{r}_2 [14],

$$\mathbf{G}_0(\mathbf{r}) = -\frac{\exp(i\omega r)}{4\pi r} [P(\omega r) (\mathbf{I} - \hat{\mathbf{r}}\hat{\mathbf{r}}) + Q(\omega r) \hat{\mathbf{r}}\hat{\mathbf{r}}] + \frac{\mathbf{I}}{3\omega^2} \delta(\mathbf{r}), \quad (8)$$

in which $(\hat{\mathbf{r}}\hat{\mathbf{r}})_{ij} = r_i r_j / r^2$, and P, Q are known functions [14].

Inside a medium, the propagator $\mathbf{G}(\mathbf{r}_1, \mathbf{r}_2)$ will be different from the free propagator. For liquids and gases we have to average this quantity over the disorder, usually performed by averaging over the positions of the scatterers [15]: $\langle \mathbf{G}(\mathbf{r}_1, \mathbf{r}_2) \rangle \equiv \mathbf{G}(\mathbf{r}_1 - \mathbf{r}_2)$. To calculate its Fourier transform $\mathbf{G}(\mathbf{k})$, it is expedient to introduce the self-energy $\Sigma(\mathbf{k})$ defined by

$$\mathbf{G}(\mathbf{k}) = \frac{1}{\omega^2 \mathbf{I} - k^2 \mathbf{I} + \mathbf{k}\mathbf{k} - \Sigma(\mathbf{k})}. \quad (9)$$

$\Sigma(\mathbf{k})$ characterizes the response of the medium and vanishes in empty space. The Green's function \mathbf{G} can be expanded into a series of scattering events that can be classified either as singly or multiply connected [15]. A singly connected event can, in contrast to a multiply connected event, be written as the product of lower-order events. The expedience of the self-energy Σ arises from the fact that it represents solely multiply connected scattering events. The dielectric function $\epsilon(\mathbf{k})$ is related to the self-energy by

$$\epsilon(\mathbf{k}) = \mathbf{I} - \frac{\Sigma(\mathbf{k})}{\omega^2}. \quad (10)$$

The widely used independent scattering approximation (ISA) amounts to keeping in Σ only the term which is lowest order in the density. This means that light can scatter from as many particles as many times as possible, but never more than once from the same particle. In addition, particle correlations are not considered within the ISA. The ISA self-energy reads

$$\Sigma^{(1)}(\mathbf{r}_1 - \mathbf{r}_2) = \rho \int d\mathbf{R}_i \mathbf{t}_i(\mathbf{r}_1, \mathbf{r}_2), \quad (11)$$

$$= -\rho \alpha \omega^2 \delta(\mathbf{r}_1 - \mathbf{r}_2) \mathbf{I}, \quad (12)$$

so that $\Sigma^{(1)}(\mathbf{k}) = -\rho \alpha \omega^2 \mathbf{I}$. The superscript in $\Sigma^{(1)}$ indicates that only the first-order terms in the density ρ are accounted for. Equation (11) implies that ISA gives for the dielectric function

$$\epsilon^{(1)} = \mathbf{I} + \rho \alpha \mathbf{I}, \quad (13)$$

which is equivalent to Eq. (1) following from response theory. To improve on this result we go beyond ISA and include particle correlations.

Let $g_m(1, \dots, m)$ be the probability distribution for observing m particles at positions $\mathbf{R}_1, \dots, \mathbf{R}_m$. These distributions can be decomposed by employing the so-called irreducible correlation functions $h_m(1, \dots, m)$ according to

$$\begin{aligned} g_1(1) &= h_1(1) = 1, \\ g_2(12) &= h_1(1)h_1(2) + h_2(12), \\ g_3(123) &= 1 + h_1(1)h_2(23) + h_1(2)h_2(13) \\ &\quad + h_1(3)h_2(12) + h_3(123), \end{aligned} \quad (14)$$

and so on. The irreducible correlation function h_m contains that part of g_m that cannot be written as a linear combination of products of correlation functions h of lower

order. As a result, the terms in the sum represent all possible partitions of the m arguments of g_m . Within the ISA, all h_m are taken to vanish for $m > 1$. Except for h_1 , the physical content and mathematical structure of the irreducible correlation functions h_m 's are highly nontrivial. Fortunately, for classical hard spheres [16] the h_m 's are required only for those coordinates in configuration space for which all m spheres overlap, to be referred to as the forbidden region. Correlation functions in this region will be denoted by a union sign: g_m^{\cup} and h_m^{\cup} . Obviously, $g_m^{\cup} = 0$ for $m > 1$, from which one finds $h_2^{\cup} = -1$, $h_3^{\cup} = +2$, and $h_4^{\cup} = -6$. The computation of high-order h_m^{\cup} becomes quickly cumbersome. The correlation functions (14) enter when the Green's function \mathbf{G} , expanded as a series of scattering events, is averaged over the particle positions. One can then unambiguously distinguish singly and multiply connected events. For the self-energy only the latter should be considered: All relevant particle correlations in \mathbf{G} are then automatically taken into account.

Within ISA, the scattering from particle i to particle j has been counted as $\mathbf{t}_i \mathbf{G}_0 \mathbf{t}_j$ in the Green's function \mathbf{G} for all possible positions of particles i and j . To account for the forbidden region up to second order in ρ , we include in Σ the following contribution:

$$\begin{aligned} \Sigma^{(2)}(\mathbf{r}_1 - \mathbf{r}_2) &= \rho^2 \int d\mathbf{r}_3 \int d\mathbf{r}_4 \int d\mathbf{R}_i \int d\mathbf{R}_j \\ &\times h_2(ij) \mathbf{t}_i(\mathbf{r}_1, \mathbf{r}_3) \cdot \mathbf{G}_0(\mathbf{r}_3 - \mathbf{r}_4) \\ &\cdot \mathbf{t}_j(\mathbf{r}_4, \mathbf{r}_2). \end{aligned} \quad (15)$$

For Rayleigh scatterers the range of $h_2(ij)$ is always smaller than the wavelength, and the dominant contribution to the integral comes from the delta function in the Green's function (8). The other contributions are of order $(\omega R)^2$ and will be neglected. Using Eq. (7) shows that $\Sigma^{(2)}(\mathbf{r}_1 - \mathbf{r}_2) = \Sigma^{(2)} \delta(\mathbf{r}_1 - \mathbf{r}_2) \mathbf{I}$ with

$$\begin{aligned} \Sigma^{(2)} &= \frac{\rho^2 \alpha^2 \omega^2}{3v^2} \int d\mathbf{R}_i \int d\mathbf{R}_j h_2(ij) \hat{\theta}_i(\mathbf{r}_1) \hat{\theta}_j(\mathbf{r}_1), \\ &= \frac{\rho^2 \alpha^2 \omega^2}{3v^2} h_2^{\cup} \int \hat{\theta}_i(\mathbf{r}_1) d\mathbf{R}_i \int \hat{\theta}_j(\mathbf{r}_1) d\mathbf{R}_j, \\ &= -\frac{\rho^2 \alpha^2 \omega^2}{3}, \end{aligned} \quad (16) \quad (17) \quad (18)$$

where $h_2(ij)$ is only required in the forbidden region. This result agrees with the LLR if the latter is expanded to second order in the density.

Subsequent contributions due to correlations turn out to obey $\Sigma^{(m)}(\mathbf{r}_1 - \mathbf{r}_2) = \Sigma^{(m)} \delta(\mathbf{r}_1 - \mathbf{r}_2) \mathbf{I}$. The third-order contribution follows from the three-particle events $\mathbf{t}_i \mathbf{G}_0 \mathbf{t}_j \mathbf{G}_0 \mathbf{t}_k$ in which particles i , j , and k overlap. By considering $\Sigma^{(2)}$ we have already taken into account the following singly connected three-particle events in \mathbf{G} : $h_2(ij) \mathbf{t}_i \mathbf{G}_0 \mathbf{t}_j \mathbf{G}_0 \mathbf{t}_k$ and $h_2(jk) \mathbf{t}_i \mathbf{G}_0 \mathbf{t}_j \mathbf{G}_0 \mathbf{t}_k$, but not yet $h_2(ik) \mathbf{t}_i \mathbf{G}_0 \mathbf{t}_j \mathbf{G}_0 \mathbf{t}_k$ and $h_3(ijk) \mathbf{t}_i \mathbf{G}_0 \mathbf{t}_j \mathbf{G}_0 \mathbf{t}_k$ which are multiply connected events. Note that the order of the

particles in the functions h_m is important. The remaining three-particle correlations to be included are thus

$$\begin{aligned} \Sigma^{(3)} &= \frac{\rho^3 \alpha^3 \omega^2}{9v^3} \int d\mathbf{R}_i \int d\mathbf{R}_j \int d\mathbf{R}_k \\ &\times [h_1(j)h_2(ik) + h_3(ijk)] \hat{\theta}_i(\mathbf{r}_1) \hat{\theta}_j(\mathbf{r}_1) \hat{\theta}_k(\mathbf{r}_1), \end{aligned} \quad (19)$$

$$= H_3 \frac{\rho^3 \alpha^3 \omega^2}{9} = \frac{\rho^3 \alpha^3 \omega^2}{9}, \quad (20)$$

in which $H_3 \equiv h_1(j)h_2^{\cup}(ik) + h_3^{\cup}(ijk) = 1$. The third-order contribution agrees again with the LLR, when the latter is expanded to third order in the density.

The combination of h_n ($n \leq m$) that appears in $\Sigma^{(m)}$, denoted by H_m , is the sum of those products that enter in multiply connected diagrams involving h , \mathbf{t} , and \mathbf{G}_0 . Inspection shows that the factorization of volume integrations effected in Eqs. (17) and (19) applies to all orders. Hence, the self-energy to all orders becomes

$$\Sigma = -\rho \alpha \omega^2 \sum_{m=0}^{\infty} H_{m+1} \left(-\frac{\rho \alpha}{3} \right)^m. \quad (21)$$

The first three orders have been determined, where $H_1 = 1$, $H_2 = -1$, and $H_3 = 1$.

Now we focus solely on the properties of H_m , and we shall prove that

$$H_m = (-1)^{m+1}. \quad (22)$$

The proof entails the generating function

$$F(\lambda) \equiv \frac{1}{1 - \sum_{i=1}^{\infty} \lambda^i H_i}. \quad (23)$$

Let us expand the generating function in powers of λ , $F(\lambda) \equiv 1 + \sum_{m=1}^{\infty} \lambda^m p_m$. The coefficient p_m is the linear combination of all possible ordered products of H_i with indices adding up to m . These terms can be associated one-to-one with the diagrams of all possible scattering events involving m particles, therefore $p_m = g_m^{\cup}$. Finally, one obtains that

$$F(\lambda) = 1 + \sum_{m=1}^{\infty} \lambda^m g_m^{\cup} = 1 + \lambda. \quad (24)$$

Using Eqs. (23) and (24) yields Eq. (22). Let us remind the reader that the proof requires infinite-order particle correlation functions.

The remarkable property that $H_{m+1} = H_{m-n} \times H_n$ shows that uncontrolled approximations, such as the one involved in the transition from Eq. (4) to Eq. (5), may turn out to give correct answers. The decoupling property of H_m looks deceptively simple. To appreciate its meaning, it is worthwhile to rewrite the H correlation functions in terms of the irreducible particle correlations h . For instance, for three-particle correlations the exact decoupling property implies $h_3^{\cup} = -3h_2^{\cup} - h_1^3$. This result can be contrasted with the well-known (Kirkwood) superposition

approximation [17] $g_3(123) = g_2(12)g_2(23)g_2(13)$, which would imply for the irreducible correlation function in the forbidden regime $h_3^U = (h_2^U)^3 + 3(h_2^U)^2$.

We have used hard-core interaction potentials. It is expected that this is not a severe limitation. Any realistic intermolecular potential will have an inner region that consists of a hard repulsive part. As most of the polarizability of the molecule will be localized within this region, our treatment can be employed for this case as well.

We can apply our microscopic theory to mixtures. To describe a mixture of type *A* and type *B* hard Rayleigh spheres, we first look at the contributions to the dielectric tensor that are linear in ρ_A and contain all orders of ρ_B . To obtain the events linear in ρ_A we just look at all *B* events in which we are allowed to replace one *t* matrix of *B* by a *t* matrix of *A*. This changes only the multiplicity of the contributions, as, for instance, the two-particle contribution $\mathbf{t}_A \mathbf{G}_0 \mathbf{t}_B$ has now become distinguishable, but equal in size, from the $\mathbf{t}_B \mathbf{G}_0 \mathbf{t}_A$ contribution. The resulting, more complicated, series can again be summed exactly and gives $\rho_A \alpha_A / (1 - \frac{1}{3} \rho_B \alpha_B)^2$, and can, for instance, be used to calculate the medium-enhanced absorption of small concentrations of *A* in *B*. In fact, it is straightforward to include all orders of ρ_A and ρ_B , and the LLR for *AB* mixtures becomes

$$\epsilon_{AB} = \mathbf{I} + \frac{\rho_A \alpha_A + \rho_B \alpha_B}{(1 - \frac{1}{3} \rho_A \alpha_A - \frac{1}{3} \rho_B \alpha_B)} \mathbf{I}, \quad (25)$$

which, if the densities are transformed into volume fractions, represents essentially the well-known Maxwell-Garnett formula.

Having established a microscopic framework that by summing a class of events generates the LLR, one can now ponder over systematic extensions. Events not included in our theory are the events in which the same scatterer is visited more than once, often called dependent scattering, as well as correlations with a range comparable to or larger than the wavelength. Dependent scattering becomes important when $\frac{1}{3} \rho \alpha \approx 1$.

Dependent scattering for two *point* particles has been considered in the static limit [9] and near an internal resonance. In the latter regime these contributions appear to become important [18]. Near internal resonances, one can consider the modification of the Einstein coefficient for spontaneous emission, where local-field concepts play an important role [19]. Recently, the Lorentz local-field corrections to nonlinear optical susceptibilities have been calculated [20]. At present, we are not able to extend our theory to include these nonlinear effects.

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Note added.—Our derivation is valid for finite frequency. The limit to zero frequency can be taken in our theory. It was pointed out to us that a derivation for precisely zero frequency actually exists [21]. In that paper, the values for the correlations functions H_m have been obtained in an alternative way. We thank B. U. Felderhof for drawing our attention to his work.

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