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Dielectric constant of a two-component granular composite: A practical scheme for calculating the pole spectrum

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A new method is presented for a systematic calculation of the effective dielectric constant ϵ_e of a granular, two-component composite material with a precisely known microscopic geometry. The basic approach is to attempt to calculate the poles and residues of ϵ_e as a function of the dielectric constants of the pure components. This calculation is reduced to an eigenvalue problem with a non-negative, self-adjoint, bounded linear operator. That problem is dealt with in two stages: First the eigenvalue problem for each of the individual, isolated grains is solved. Then the general eigenvalue problem for the entire composite is expanded in terms of the individual grain eigenfunctions, and in this way it becomes a matrix eigenvalue problem. The method is applied to a number of physical systems, including a general periodic composite, and a simple-cubic lattice of identical spheres. The results include some new predictions concerning additional optical resonances which should appear in periodic or quasiperiodic metal-insulator granular composites.

I. INTRODUCTION AND SUMMARY

In a previous article, we discussed the general analytical properties of the complex dielectric constant of a composite medium ϵ_e viewed as a function of the complex dielectric constants of the pure, homogeneous constituents ϵ_1, ϵ_2, etc.[1] This was done by introducing a characteristic geometric function, which depends only on ratios of the ϵ_i's. In the case of a two-component or two-phase medium, this function is defined by

$$h \equiv \epsilon_1/\epsilon_2 ,$$

$$m(h) \equiv \epsilon_e/\epsilon_2 . \qquad (1.1)$$

For a specific, finite sample of the composite, this function was shown to be analytic everywhere (including the point $h=\infty$) except for a finite number of simple poles with negative residues on the negative real axis of h. Therefore, a knowledge of the poles and the residues completely determines the function $m(h)$.

In this article we will develop a general method for calculating these poles and their residues. The basic idea is to recast the problem as an eigenvalue problem of a linear, non-negative, self-adjoint operator—the eigenvalues being the poles of $m(h)$. Once this has been done, one can solve the problem in stages: for a composite made of grains ϵ_1 embedded in a host material ϵ_2, we can first solve the eigenvalue problem for each isolated grain separately. We then expand the eigenfunctions of the entire composite using the eigenfunctions of the individual grains. In this way, we get an eigenvalue problem with an Hermitian matrix whose elements can be represented in the form of overlap integrals involving eigenfunctions of two grains.

When the general formalism thus obtained is applied to the problem of two grains, and to the problem of a periodic lattice of identical grains, the results are somewhat surprising: when two identical grains are considered, it is found, as one might have guessed, that the mutual influence or interaction between the grains splits each of the doubly degenerate eigenvalues into two distinct eigenvalues. But the residues or weights are not equally divided: one of the split poles carries all of the weight, while the other has a vanishing residue! Similarly, the interactions between the grains of a periodic composite cause every eigenvalue of the individual grain to be broadened into a quasi continuous band, in which the different states are characterized by a wave vector $\vec{k}$ in the first Brillouin zone of the appropriate reciprocal lattice. But again the weights are far from being uniformly distributed: in each band, all the weight is concentrated in the $\vec{k}=0$ eigenvalue. Therefore, the spectrum of poles contributing to $m(h)$ for such a composite is discrete, as it was in the case of the isolated grain, with every pole having a finite, rather than an infinitesimal weight. In order to get a spectrum of poles with weights that are smeared quasicontinuously one presumably needs to have a disordered composite.

The outline of this article is as follows. Section II shows how the calculation of $m(h)$ can be reduced to the problem of finding all the eigenvalues and eigenfunctions of a certain linear operator. In Section III we apply this method to a series of problems, starting with an isolated spherical inclusion; a system of interacting inclusions; a pair of identical or similar (i.e., related to each other by a uniform dilatation) inclusions; a general periodic array of inclusions; and finally a simple cubic array of identical spheres. A prediction is made about the existence of hitherto unobserved additional resonances in the optical properties of certain metal-insulator granular composites. In Sec. IV the effect of disorder on these results is discussed qualitatively. Finally, in the Appendix we calculate the matrix elements (or overlap integrals) for a pair of spherical inclusions.

II. GENERAL THEORY OF THE POLE SPECTRUM

The poles of $m(h)$ were shown in Ref. 1 to be eigenvalues of the following linear homogeneous boundary-value problem for the electrostatic potential ψ in a parallel plate condenser with perpendicular walls, filled with the composite material under discussion

$$\begin{aligned} &\vec{\nabla}\cdot[(h\theta_1+\theta_2)\vec{\nabla}\psi]=0\,,\\ &\psi=0 \quad\text{on the condenser plates}\,, \qquad (2.1)\\ &\frac{\partial\psi}{\partial n}=0 \quad\text{on the condenser walls}\,. \end{aligned}$$

Here $\partial\psi/\partial n$ denotes the normal derivative, while $\theta_i(\vec{r})=1$ if $\vec{r}$ is inside phase i material and $\theta_i(\vec{r})=0$ otherwise.

It was shown in Ref. 1 that all the eigenvalues h_α of (2.1) are real and negative. It is clear that an eigenfunction ψ_α describes a situation where a spurious electric field exists inside the condenser without any external field or potential difference being applied. In order to find an expression for the residue of h_α, we focus attention on the reciprocal function $\tilde{m}(\tilde{h})\equiv 1/m(h)$, where $\tilde{h}\equiv 1/h$. Obviously, near a pole of $m(h)$ we can write

$$\begin{aligned} &m(h)\cong -A_\alpha/(h-h_\alpha)\,,\\ &\tilde{m}'(\tilde{h}_\alpha)=h_\alpha^2/A_\alpha\,, \qquad (2.2) \end{aligned}$$

since $\tilde{h}_\alpha$ is a regular point (in fact, a zero) of $\tilde{m}(\tilde{h})$. The derivative $\tilde{m}'(\tilde{h})$ was evaluated in Ref. 1 in terms of the solution of the following inhomogeneous boundary-value problem defined on the same condenser as (2.1)

$$\begin{aligned} &\vec{\nabla}\cdot[(h\theta_1+\theta_2)\vec{\nabla}\phi]=0\,,\\ &\phi=0 \quad\text{on one plate,}\\ &\phi=\text{ const. on the other plate,} \qquad (2.3)\\ &\frac{\partial\phi}{\partial n}=0 \quad\text{on the walls,}\\ &\frac{1}{S}\int(h\theta_1+\theta_2)\frac{\partial\phi}{\partial n}dS=1\,. \end{aligned}$$

This problem describes the situation where a given total charge is placed upon the plates of the condenser. The surface integral appearing in (2.3), which can be taken over any one of the condenser plates, describes that charge. The condition on the surface charge could also have been replaced by

$$\frac{1}{V}\int(h\theta_1+\theta_2)\frac{\partial\phi}{\partial z}dV=1\,, \qquad (2.4)$$

where the integration is now over the entire volume V and where the z axis has been chosen to be perpendicular to the plates.

As was noted in Ref. 1, the eigenfunction ψ_α associated with a pole of $m(h)$ is proportional to the solution of (2.3), and hence we can write [see Eq. (II. 16) of Ref. 1]

$$\begin{aligned} \tilde{m}'(\tilde{h}_\alpha)&=\frac{1}{V}\int\theta_1(h_\alpha\theta_1+\theta_2)^2(\vec{\nabla}\phi)^2dV\\ &=h_\alpha^2\frac{(1/V)\int\theta_1(\nabla\psi_\alpha)^2}{[(1/V)\int(h_\alpha\theta_1+\theta_2)\partial\psi_\alpha/\partial z\,dV]^2}\,. \qquad (2.5) \end{aligned}$$

From this, we can immediately obtain an expression for the residue A_α.

In what follows, it will be more convenient to use an alternative form of the characteric geometric function, namely,[1]

$$\begin{aligned} &s\equiv 1/(1-h)\\ &F(s)\equiv 1-m\equiv\sum_\alpha\frac{B_\alpha}{s-s_\alpha}\,, \qquad (2.6) \end{aligned}$$

where the residues B_α are all on the semiclosed segment $[0,1)$. Using (2.5) and (2.2), we can write

$$B_\alpha=M_\alpha^2 \qquad (2.7\text{a})$$

$$\begin{aligned} M_\alpha&=s_\alpha\frac{(1/V)\int(h_\alpha\theta_1+\theta_2)(\partial\psi_\alpha/\partial z)\,dV}{[(1/V)\int\theta_1(\vec{\nabla}\psi_\alpha)^2dV]^{1/2}}\\ &=\frac{(1/V)\int\psi_\alpha(\partial\theta_1/\partial z)dV}{[(1/V)\int\theta_1(\vec{\nabla}\psi_\alpha)^2dV]^{1/2}}\,, \qquad (2.7\text{b}) \end{aligned}$$

where integration by parts of the numerator as well as the boundary conditions on ψ_α have been used to get the final result. We note that M_α is proportional to the total charge that is present on the plates in the given eigenmode of the condenser, that it can be negative as well as positive, and that it can vanish for some of these modes. (As we shall see below, it must in fact vanish for most of the eigenmodes.) We will call M_α the amplitude of the pole or eigenvalue h_α, in order to distinguish it from the residue or weight B_α.

In order to make further progress, we recast the eigenvalue problem of (2.1) in the form of a linear integral equation with a self-adjoint kernel. This is done with the help of Green's function for Laplace's equation with boundary conditions on the surface of the condenser

$$\begin{aligned} &\nabla^2 G(\vec{r}\,|\,\vec{r}\,') = -\,\delta^d(\vec{r}-\vec{r}\,') \\ &G = 0 \quad \text{on both plates,} \\ &\frac{\partial G}{\partial n} = 0 \quad \text{on the walls.} \end{aligned} \tag{2.8}$$

Using the fact that $\theta_1 + \theta_2 \equiv 1$, we can write the differential equation of (2.1) in the form

$$\nabla^2\psi = \frac{1}{s}\vec{\nabla}\cdot(\theta_1\vec{\nabla}\psi)\,. \tag{2.9}$$

We can then use Green's function to "solve" this equation in the form

$$\begin{aligned} s\psi(\vec{r}) &= -\int G(\vec{r}\,|\,\vec{r}\,')\vec{\nabla}'\cdot[\theta_1(\vec{r}\,')\vec{\nabla}'\psi(\vec{r}\,')]\,dV' \\ &= \int \theta_r(\vec{r}\,')\vec{\nabla}'G(\vec{r}\,|\,\vec{r}\,')\cdot\vec{\nabla}\psi(\vec{r}\,')dV'\,, \end{aligned} \tag{2.10}$$

where we integrated by parts and used the boundary conditions on G and on ψ to get the final result. The right-hand side of (2.10) is a linear functional of ψ which we denote by $\hat{G}\psi$. If we now define the scalar product of any two real functions by

$$(\phi,\psi) \equiv \int \theta_1\vec{\nabla}\phi\cdot\vec{\nabla}\psi\,dV\,, \tag{2.11}$$

then we can show that $\hat{G}$ is a self-adjoint, nonnegative, bounded linear operator. In order to show the self-adjoint property, we only need to use the fact that G is real and satisfies[2]

$$G(\vec{r}\,|\,\vec{r}\,') = G(\vec{r}\,'\,|\,\vec{r})\,, \tag{2.12}$$

and write for any two functions ϕ, ψ

$$\begin{aligned} (\psi,\hat{G}\psi) &= \int dV\int dV'\,\theta_1(\vec{r})\theta_1(\vec{r}\,')\sum_{\alpha,\beta}\frac{\partial G(\vec{r}\,|\,\vec{r}\,')}{\partial r_\alpha \partial r'_\beta} \\ &\qquad\qquad \times\frac{\partial\phi(\vec{r})}{\partial r_\alpha}\frac{\partial\psi(\vec{r}\,')}{\partial r'_\beta} \\ &= (\hat{G}\phi,\psi)\,. \end{aligned} \tag{2.13}$$

That Green's function is real results from the fact that the inhomogeneous differential equation (2.8) is real. The non-negativity and boundedness of $\hat{G}$ follow from the fact that the spectrum of eigenvalues is contained in the semiclosed segment $[0, 1)$.

While it is possible to conduct the entire discussion in terms of the real scalar product of (2.11), because the eigenfunctions of $\hat{G}$ can all be chosen to be real, it is often convenient to allow complex solutions. The reasons for this are similar to those that sometimes lead us to prefer complex eigenfunction solutions of Schrödinger's equation even when the Hamiltonian is real. We therefore generalize the definition of the scalar product to include complex functions

$$(\phi,\psi) \equiv \int \theta_1\vec{\nabla}\phi^*\cdot\vec{\nabla}\psi\,dV\,. \tag{2.14}$$

The norm of a function will be denoted, as usual, by $\|\psi\|$

$$\|\psi\| \equiv (\psi,\psi)^{1/2}\,. \tag{2.15}$$

We must now also allow the amplitudes M_α to be complex, and we must therefore replace (2.7a) and (2.7b) by

$$B_\alpha = |M_\alpha|^2\,, \tag{2.16a}$$

$$M_\alpha \equiv \frac{\int\psi_\alpha(\partial\theta_1/\partial z)\,dV}{V^{1/2}\|\psi_\alpha\|}\,. \tag{2.16b}$$

As a consequence of the above discussion, the eigenvalue problem (2.1) can be reformulated as the eigenvalue problem of the integral operator $\hat{G}$

$$\hat{G}\psi = s\psi\,. \tag{2.17}$$

The boundary conditions of (2.1) are automatically satisfied by any eigenfunction solution of (2.17), due to the properties of Green's function G. This formulation now enables the entire machinery developed for handling such eigenvalue problems to be used for the evaluation of the poles of $m(h)$ and their amplitudes.

In particular, we can immediately conclude that the eigenfunctions of $\hat{G}$ form a complete orthogonal set with respect to the scalar product of (2.14). Thus, any piecewise continuously differentiable function can be expanded inside the phase-1 volume in a series of these functions. For this to be true, the total number of eigenfunctions must be infinite, which would seem to contradict our earlier assertion that the total number of poles of $m(h)$ is finite for a finite composite. The only way to reconcile these two correct statements is by having only a finite number of nonvanishing amplitudes M_α. Thus, most of the eigenmodes of $\hat{G}$ must involve a spurious electric field in the system that results in no charge being accumulated on the condenser plates. We will see an example of this in Sec. III.

Before closing this section, we will note a certain symmetry property of the eigenvalue problem: if two composite systems have similar geometries, i.e., if one can be obtained from the other by a simple uniform dilatation $\vec{r}\,' = \lambda^{-1}\vec{r}$, then the eigenfunctions, the norms, the θ_1 function, and Green's function for the two systems are also obtained from each other by a simple transformation

$$\begin{aligned} &\psi'_\alpha(\vec{r}\,') = \psi_\alpha(\vec{r}) = \psi_\alpha(\lambda\vec{r}\,')\,, \\ &\|\psi'_\alpha\|^2 = \lambda^{2-d}\|\psi_\alpha\|^2\,, \\ &\theta'_1(\vec{r}\,') = \theta_1(\lambda\vec{r}\,')\,, \\ &G'(\vec{r}\,'\,|\,\vec{r}\,'_0) = \lambda^{d-2}G(\lambda\vec{r}\,'\,|\,\lambda\vec{r}\,'_0)\,, \end{aligned} \tag{2.18}$$

where d is the dimensionality of space. The eigenvalues are invariant under this transformation, and are the same in both systems, and the same result is obtained for the amplitudes by using (2.18) in (2.16b), as well as the fact that $V' = \lambda^{-d}V$.

There are cases, however, where the total volume is so much greater than the volume of phase 1, that one may perform the dilatation transformation only on that phase and still use Eq. (2.18) to describe the changes that occur within that phase. In such a case, the $V^{1/2}$ factor appearing explicitly in the denominator of (2.16b) remains unchanged and the amplitudes are altered, becoming

$$M'_\alpha = \lambda^{-d/2} M_\alpha \,. \tag{2.19}$$

Finally, we should note that an equation similar to our Eq. (2.10) has been used in the past by Englman and Ruppin[3] to calculate the optical lattice modes in a finite crystal. The novel feature of our paper, which will appear in Sec. III, is the use of that equation to calculate the modes of a collection of many small grains.

III. APPLICATION TO SOME SPECIFIC GEOMETRIES

A. Single spherical inclusion

The first problem we will discuss is that of a single spherical inclusion ϵ_1, situated at the center of a very large cylindrical condenser filled with ϵ_2 host material. We introduce polar coordinates with the origin at the center of the sphere and the polar axis along the cylinder axis. The potential function can be expanded in a series of the functions

$$r^l Y_{lm}(\Omega), \quad r^{-l-1} Y_{lm}(\Omega), \tag{3.1}$$

where Y_{lm} is a spherical harmonic.

If we take the boundaries of the condenser to be infinitely far away from the spherical inclusion, then inside the sphere we will have only the functions $r^l Y_{lm}$, while outside the sphere we will have only the functions $r^{-l-1} Y_{lm}$. Furthermore, the $l = 0$ function will be absent outside, because its appearance would have required the presence of a total nonvanishing charge inside the sphere. The boundary conditions on the plates and walls are thus automatically satisfied. The differential equation of (2.1) is also automatically satisfied by these functions, except at the surface of the sphere, where ψ as well as $\epsilon\partial\psi/\partial n$ must undergo no discontinuity upon passing through the surface.

These continuity conditions lead to the following series of eigenfunctions and eigenvalues

$$\psi_{lm}(\vec{r}) = \begin{cases} r^l Y_{lm}(\Omega), & \text{for } r < a_0 \\ \dfrac{a_0^{2l+1}}{r^{l+1}} Y_{lm}(\Omega), & \text{for } r > a_0, \\ l = 1, 2 \ldots; \quad -l \leqslant m \leqslant l \,, \end{cases} \tag{3.2}$$

$$h_{lm} = h_l = -\frac{l+1}{l}, \quad s_{lm} = s_l = \frac{1}{1-h_l} = \frac{l}{2l+1}, \tag{3.3}$$

where a_0 is the radius of the spherical inclusion. Note that the mutual orthogonality of these functions with respect to the scalar product of (2.11) is different from the usual orthogonality property of the spherical harmonies

$$\begin{aligned}(\psi_{lm}, \psi_{l'm'}) &= \int_{r<a_0} r^2 dr\, d\Omega\, \vec{\nabla}(r^l Y^*_{lm}) \cdot \vec{\nabla}(r^{l'} Y_{l'm'}) \\ &= \int_{r=a_0} r^2 d\Omega\, r^l Y^*_{lm} \frac{\partial}{\partial r}(r^{l'} Y_{l'm'}) \\ &= l' a_0^{l+l'+1} \int d\Omega\, Y^*_{lm} Y_{l'm'} \\ &= l a_0^{2l+1} \delta_{ll'} \delta_{mm'} \,. \end{aligned} \tag{3.4}$$

Note also that this is an example where most of the eigenfunctions are in fact complex.

The amplitudes M_{lm} can now be calculated by using (2.16b), and noting that

$$\begin{aligned}\frac{\partial\theta_1}{\partial z} = \delta(r-a_0)\frac{\partial r}{\partial z} &= \cos\theta\, \delta(r-a_0) \\ &= (\tfrac{4}{3}\pi)^{1/2} Y_{10}(\Omega) \delta(r-a_0) \,. \end{aligned} \tag{3.5}$$

Because of the orthogonality of the spherical harmonics, we find that the only nonzero weight is

$$\begin{aligned} M_{10} &= \frac{(a_0^3/V)(\frac{4}{3}\pi)^{1/2} \int d\Omega\, Y_{10}^2}{[(3/4\pi V)\int_{r<a_0} (\vec{e}_z)^2 dV]^{1/2}} \\ &= \frac{v/V}{(v/V)^{1/2}} = \left(\frac{v}{V}\right)^{1/2}, \end{aligned} \tag{3.6}$$

where $v \equiv 4\pi a_0^3/3$ is the volume of the sphere. This is in agreement with the sum rule, proved in Ref. 1, which states that the total sum of all residues is equal to the volume fraction of phase-1 material. Of course, M_{10} itself also vanishes in the limit $V \to \infty$ which we have been using. But the other amplitudes vanish "exactly" in this approximation. What that must mean is that if we had made a more honest calculation on a sphere within a large but finite cylinder, we would have found an additional finite number of poles with very small but nonzero amplitudes. The total number of these poles probably increases to infinity as $V \to \infty$, but the sum of all their weights must tend to 0 faster than v/V.

B. System of interacting grains

Suppose that the composite is made of a number of grains (inclusions) ϵ_1 embedded in the ϵ_2 host, and that we know the eigenfunctions and eigenvalues of each grain separately in the same host. When they are present together, the grains will influence each other or "interact," so that the spectrum of the entire system will not be simply the sum of the spectra of the individual grains. This is reminiscent of the problem of the energy levels of a collection of atoms whose individual levels are altered as a result of mutual interactions.

In order to take advantage of the available information, we will try to expand the potential ψ in terms of the potential eigenfunctions $\psi_{a\alpha}$ of the individual grains, denoted by the index a. Clearly, we cannot hope to expand an arbitrary function everywhere in terms of these functions. In fact, it is clear that any function which vanishes inside all of the grains will be orthogonal to all of the eigenfunctions. We therefore attempt to expand $\psi(\vec{r})$ in this way only for points $\vec{r}$ that are essentially inside one of the grains

$$\theta_1^+(\vec{r})\psi(\vec{r}) = \sum_{a\alpha} \frac{A_{a\alpha}\cdot\theta_a^+(\vec{r})\psi_{a\alpha}(\vec{r})}{\|\psi_{a\alpha}\|} \tag{3.7}$$

Here we have introduced the function $\theta_a^+(\vec{r})$, which is equal to 1 if $\vec{r}$ is either inside the grain a, or at most an infinitesimal distance away outside it, and is equal to 0 otherwise. The function θ_1^+ is defined in a similar way by considering all of the ϵ_1 grains, i.e.,

$$\theta_1^+ = \sum_a \theta_a^+ . \tag{3.8}$$

We now substitute (3.7) into the eigenvalue equation (2.10) or (2.17) noting that, by (3.8), we can write $\hat{G}$ as a sum of individual grain operators $\hat{G}_a$. When this sum operates on (3.7), every operator $\hat{G}_a$ selects only those terms that refer to its own grain. By choosing the region where $\theta_a^+ \neq 0$ to be infinitesimally larger than the actual grain itself, we have ensured that $\vec{\nabla}\theta_a^+$ vanishes everywhere inside the grain. Thus we obtain

$$\begin{aligned}\hat{G}\psi = \hat{G}(\theta_1^+\psi) &= \sum_{a\alpha} \frac{A_{a\alpha}\hat{G}_a(\theta_a^+\psi)_{a\alpha}}{\|\psi_{a\alpha}\|} \\ &= \sum_{a\alpha} \frac{A_{a\alpha}\hat{G}_a\psi_{a\alpha}}{\|\psi_{a\alpha}\|} = \sum_{a\alpha} \frac{A_{a\alpha}s_{a\alpha}\psi_{a\alpha}}{\|\psi_{a\alpha}\|} ,\end{aligned} \tag{3.9}$$

$$s\theta_1^+\psi = \sum_{a\alpha} \frac{A_{a\alpha}\, s\theta_a^+\psi_{a\alpha}}{\|\psi_{a\alpha}\|} . \tag{3.10}$$

We can equate (3.9) and (3.10) for $\vec{r}$ inside any one of the ϵ_1 grains. We then take the scalar product of the resulting equation with any individual grain eigenfunction $\psi_{b\beta}$. When we apply this procedure to (3.10), the single term $b\beta$ is selected, but when we apply it to (3.9), we are usually left with the entire sum

$$sA_{b\beta}\|\psi_{b\beta}\| = \sum_{a\alpha} A_{a\alpha}s_{a\alpha} \int \frac{\theta_b \vec{\nabla}\psi_{b\beta}^* \cdot \vec{\nabla}\psi_{a\alpha}\, dV}{\|\psi_{a\alpha}\|} . \tag{3.11}$$

This is a matrix eigenvalue equation which can be rewritten as

$$(s - s_{a\alpha})A_{a\alpha} = \sum_{\substack{b\beta \\ b\neq a}} Q_{a\alpha,b\beta}A_{b\beta} , \tag{3.12}$$

where

$$Q_{a\alpha,b\beta} \equiv \frac{s_{b\beta}\int \theta_a \vec{\nabla}\psi_{a\alpha}^* \cdot \vec{\nabla}\psi_{b\beta}\, dV}{\|\psi_{a\alpha}\|\,\|\psi_{b\beta}\|} \tag{3.13}$$

$$\begin{aligned}&= \|\psi_{a\alpha}\|^{-1}\|\psi_{b\beta}\|^{-1} \\ &\quad \times \int dV \int dV'\, \theta_a(\vec{r}) \\ &\quad \times \theta_b(\vec{r}\,') \sum_{\mu,\nu} \frac{\partial^2 G(\vec{r}\,|\,\vec{r}\,')}{\partial r_\mu \partial r'_\nu} \\ &\quad \times \frac{\partial\psi_{a\alpha}^*(\vec{r})}{\partial r_\mu}\frac{\partial\psi_{b\beta}(\vec{r}\,')}{\partial r'_\nu} .\end{aligned} \tag{3.14}$$

Once Eq. (3.12) has been solved and we know an eigenvalue $s^{(i)}$ and the appropriate eigenvector $A_{a\alpha}^{(i)}$, we can calculate the amplitude $M^{(i)}$ by substituting (3.7) into (2.16b). In this way we get

$$M^{(i)} = \left(\sum_{a\alpha} A_{a\alpha}^{(i)} M_{a\alpha}\right) \Big/ \left(\sum_{a\alpha} |A_{a\alpha}^{(i)}|^2\right)^{1/2} . \tag{3.15}$$

We note that it is possible to have eigenstates of the individual grain that do not contribute a pole to $m(h)$ of the isolated grain, but which nevertheless play a role in determining the poles of the multigrain system. For example, while only $M_{10} \neq 0$ in the case of an isolated spherical inclusion, there are nonzero overlap integrals between that state and many other states of other grains. Because of this a system of interacting spherical grains will have many poles, in contrast with the single pole of an isolated spherical grain.

The scheme we have developed in this subsection can be useful if a reasonable approximation can be obtained by truncating the infinite set of equations (3.12) or a simple transformation thereof. In the following subsections we will investigate this question for the case of two identical grains, and for a periodic lattice of grains. An approach that is somewhat reminiscent of ours has recently been proposed by Clippe *et al.*[4] However, they restrict the applicability of their discussion by assuming that the different grains in the composite interact only through dipole-dipole forces.

C. Two identical or similarly shaped small grains

In this case, the unperturbed eigenvalues are all at least twofold degenerate, since the spectra of the two separate grains are identical. Therefore, even if the grains are far away and interact only very weakly, the perturbed eigenstates will be very different from the unperturbed states. If we neglect the interactions with other, nondegenerate eigenstates, and concentrate only on the degenerate pair ψ_{a0}, ψ_{b0}, the resulting equations for the expansion coefficients are

$$\begin{aligned} A_{a0}(s-s_0) &= A_{b0}C\,, \\ A_{b0}(s-s_0) &= A_{a0}C^*\,, \end{aligned} \tag{3.16}$$

where

$$C \equiv s_0 \frac{\int \theta_a \vec{\nabla}\psi^*_{a0}\cdot\vec{\nabla}\psi_{b0}\,dV}{\|\psi_{a0}\|\,\|\psi_{b0}\|}\,. \tag{3.17}$$

The solution of this little eigenvalue problem is of course

$$s_\pm = s_0 \pm |C| \tag{3.18}$$

$$A_{b0} = \pm(C/|C|)A_{a0}\,. \tag{3.19}$$

As we might have expected, the unperturbed eigenvalue is split symmetrically, while the eigenstates are the symmetric and antisymmetric linear combination of the unperturbed states.

In order to investigate what the new amplitudes are, we assume $C>0$, and use (3.15) and (3.19) to get

$$M = \frac{A_{a0}(M_{a0}\pm M_{b0})}{(2A^2_{a0})^{1/2}} = \frac{M_{a0}\pm M_{b0}}{\sqrt{2}}\,, \tag{3.20}$$

where we chose A_{a0} to be real and positive. We then use (2.19) to note that, for similarly shaped and oriented grains, the amplitude is proportional to the square root volume of the grain. Thus for identical grains the upper (symmetric) eigenvalue s_+ carries the combined weights of the unperturbed states—its residue is $M^2_+ = 2M^2_0$—while the lower (antisymmetric) eigenvalue s_- carries zero weight. On the other hand, if one of the grains is much larger than the other one, then the weights of s_+ and s_- will be roughly the same, and equal to half of the weight of the unperturbed pole.

One can understand these results on physical grounds in the following way: each of the unperturbed eigenstates ψ_{a0}, ψ_{b0} builds up a spurious charge on the condenser plates whose magnitude is proportional to the amplitude of the corresponding individual grain pole. In the symmetric combination of the eigenstates, the charges produced on a given condenser plate by the two grains have equal signs, and they add up to a greater combined charge (there is some interference however—since the total amplitude for the case of identical grains is $\sqrt{2}M_0$ and not $2M_0$). In the antisymmetric combination, by contrast, the two grains contribute opposite charges to a given plate, and the mutual cancellation results in a zero combined charge for the case of identical grains. By contrast, when the grains are very unequal, the larger one always dominates the spurious charge produced on the plates in both the symmetric and antisymmetric combinations. The weights of s_+ and s_- are then roughly equal.

D. Periodic lattice of identical grains

In this case the eigenstates and eigenvalues of all the grains are identical. Consequently the matrix element $Q_{a\alpha,b\beta}$ of (3.13) depends on the position vectors of the grains $\vec{a}$ and $\vec{b}$ only through their difference $\vec{a}-\vec{b}$. According to Bloch's theorem, the eigenvectors of (3.12) can always be written in the form

$$A_{a\alpha}(\vec{k}) \equiv A_\alpha(\vec{k})e^{i\vec{k}\cdot\vec{a}}\,. \tag{3.21}$$

When this is substituted in (3.12) we get the following equation for $A_\alpha(\vec{k})$

$$(s-s_\alpha)A_\alpha(\vec{k}) = \sum_\beta \tilde{Q}_{\alpha\beta}(\vec{k})A_\beta(\vec{k})\,, \tag{3.22}$$

where

$$\tilde{Q}_{\alpha\beta}(\vec{k}) \equiv \sum_{\vec{a}-\vec{b}\neq 0} Q_{a\alpha,b\beta}e^{-i\vec{k}\cdot(\vec{a}-\vec{b})}\,. \tag{3.23}$$

The Fourier-transormed matrix elements $Q_{\alpha\beta}(\vec{k})$ and the eigenvectors $A_\alpha(\vec{k})$ satisfy

$$\begin{aligned} \tilde{Q}^*_{\alpha\beta}(\vec{k}) &= \tilde{Q}_{\beta\alpha}(\vec{k}) = \tilde{Q}_{\alpha\beta}(-\vec{k})\,, \\ A^*_\alpha(\vec{k}) &= A_\alpha(-\vec{k})\,. \end{aligned} \tag{3.24}$$

The eigenfunction corresponding to an eigenvalue $s_i(\vec{k})$ is given by

$$\begin{aligned} \theta^+_1(\vec{r})\psi_{ik}(\vec{r}) &= \sum_{\alpha a} \theta^+_a(\vec{r})A^{(i)}_\alpha(\vec{k})e^{i\vec{k}\cdot\vec{a}}\psi_{a\alpha}(\vec{r})/\|\psi_{a\alpha}\| \\ &= \sum_\alpha A^{(i)}_\alpha(\vec{k})\phi_{\alpha k}(\vec{r}) \end{aligned} \tag{3.25}$$

for $\vec{r}$ inside phase-1 material, where

$$\phi_{\alpha k}(\vec{r}) \equiv \sum_a \frac{\theta^+_a(\vec{r})e^{i\vec{k}\cdot\vec{a}}\psi_{a\alpha}(\vec{r})}{\|\psi_{a\alpha}\|}\,. \tag{3.26}$$

The amplitude of the pole at $s_i(\vec{k})$ can be found by substituting (3.21) into (3.15) and recalling that $M_{a\alpha} = M_\alpha$. In this way we find

$$M_i(\vec{k}) = \frac{N\delta_{k,0}\sum_\alpha A^{(i)}_\alpha(0)M_\alpha}{(N\sum_\alpha |A^{(i)}_\alpha(0)|^2)^{1/2}}\,, \tag{3.27}$$

where N is the total number of grains or unit cells in the periodic lattice. The last result is the natural extension of a previous result for the two-grain system: all of the weight is concentrated at the discrete $\vec{k}=0$ poles, even though there is a quasicontinuous band of eigenvalues $s_i(\vec{k})$ for every index i. This result contradicts some statements that have appeared in the literature recently, according to which the interactions between grains in such a system must cause a broadening of the single sharp resonance (i.e., pole) that appears in the Clausius–Mossotti equation for the dielectric constant.[5] We find that in a periodic system, these interactions can only bring about the appearance of additional sharp resonances in the complex dielectric constant. We expect a broadening of the pole spectrum only as the result of disorder, e.g., deviations from perfect periodicity or variations in the sizes and shapes of the grains.

E. Simple-cubic lattice of identical spheres

This is a problem that was discussed a long time ago in a historic paper by Lord Rayleigh,[6] who developed what is essentially an expansion of $1/F(s)$ in powers of the volume fraction p_1 of ϵ_1. We are now in a position to do a better job, especially in the neighborhood of the poles and when the volume fraction is not small. We note in this connection that although by considering $1/F$ rather than F the poles are changed into zeros, the troubles do not disappear. This is so because the zeros of F, of which there is one between every pair of neighboring poles, are changed into poles of $1/F$.

The overlap integral between the eigenstates ψ_{0lm} and $\psi_{bl'm'}$ of two spheres with radii a_0, a_1, situated at the origin and at a site $\vec{b}$, respectively, is worked out in the Appendix. The resulting matrix element is given by

$$\begin{aligned}Q_{0lm,bl'm'} &\equiv \frac{s_{bl'm'}\int \theta_0 \vec{\nabla}\psi^*_{0lm}\cdot\vec{\nabla}\psi_{bl'm'}\,dV}{\|\psi_{0lm}\|\;\|\psi_{bl'm'}\|}\\ &= (-1)^{l'+m'}\frac{a_0^{l+1/2}a_1^{l'+1/2}}{b^{l+l'+1}}\left(\frac{ll'}{(2l+1)(2l'+1)}\right)^{1/2}\\ &\quad\times\frac{(l+l'+m-m')!}{[(l+m)!\,(l-m)!\,(l'+m')!\,(l'-m')!]^{1/2}}\\ &\quad\times e^{i\phi_b(m'-m)}P_{l'+l}^{(m'-m)}(\cos\theta_b),\end{aligned}\qquad(3.28)$$

where b, θ_b are the polar coordinates of the vector $\vec{b}$, while $P_l^{(m)}(u)$ is an associated Legendre polynomial, defined by[7]

$$\begin{aligned}P_l^{(m)}(u) &= (1-u^2)^{m/2}\frac{d^mP_l(u)}{du^m},\quad\text{for } m>0,\\ P_l^{(-m)}(u) &= (-1)^m\frac{(l-m)!}{(l+m)!}P_l^{(m)}(u)\,.\end{aligned}\qquad(3.29)$$

When these matrix elements are summed over the sites of a simple-cubic lattice, some simplifications occur: The sum vanishes unless $m-m'$ is an integral multiple of 4, and unless $l+l'$ is even. When $l+l'>2$, the sum will be absolutely convergent. But for $l+l'=2$, which occurs only when $l=l'=1$, $m=m'=0$, this is not the case and care must be taken to perform the sum on distant sites correctly. The sum is then over a set of potential fields created at the origin by dipole sources at all the other lattice sites. This is the same type of sum that is encountered in calculating the Lorentz local field at the origin, and can be dealt with in the same way: The sum is divided into a "near-field contribution" resulting from all the sources within a large but finite sphere around the origin, and a "far-field contribution" resulting from all the sources outside that sphere. The far-field contribution is calculated by replacing the discrete dipole sources by a uniform polarization density, while the near-field contribution must be calculated exactly. In the cubic case it vanishes.

We have seen that in the case of an isolated sphere, the only nonzero amplitude is $M_{10} = (v/V)^{1/2}$. Therefore, when the lattice is not too densely packed with spheres (i.e., $p_1 \ll 1$), only states that couple directly to $\psi_{lm}=\psi_{10}$ with a small negative power of b are important. Thus, to lowest order in p_1, we only need to consider the state ψ_{30}. We are thus left with a 2×2 matrix eigenvalue problem

$$s\begin{pmatrix}A_1\\ A_3\end{pmatrix} = \begin{pmatrix}\frac{1}{3}(1-p_1) & -(4/\sqrt{7})\Sigma_4 p_1^{5/3}\\ -(4/\sqrt{7})\Sigma_4 p_1^{5/3} & \frac{3}{7}-\frac{60}{7}\Sigma_6 p_1^{7/3}\end{pmatrix}\begin{pmatrix}A_1\\ A_3\end{pmatrix},\qquad(3.30)$$

where

$$\Sigma_n p_1^{(n+1)/3} \equiv \sum_{\vec{b}\neq 0}\left(\frac{a_0}{b}\right)^{n+1}P_n(\cos\theta_b)\,,\qquad(3.31)$$

and the sum is over all the sites of a simple-cubic lattice.

We now solve this equation to get the eigenvalues, eigenvectors, and pole amplitudes by means of standard perturbation techniques. The eigenvalues and pole amplitudes are given by

$$\begin{aligned}s^{(1)} &\cong s_1 + \frac{|Q_{30,10}|^2}{s_1-s_3} = \tfrac{1}{3}(1-p_1) - 24\Sigma_4^2 p_1^{10/3},\\ s^{(3)} &\cong s_3 + \frac{|Q_{10,30}|^2}{s_3-s_1}\\ &= \tfrac{3}{7} - \tfrac{60}{7}\Sigma_6 p_1^{7/3} + 24\Sigma_4^2 p_1^{10/3},\\ M^{(1)} &\cong \frac{N^{1/2}M_{10}}{(1+|Q_{10,30}|^2/(s_1-s_3)^2)^{1/2}}\\ &\cong (p_1 - 252\Sigma_4^2 p_1^{13/3})^{1/2},\\ M^{(3)} &\cong N^{1/2}M_{10}\frac{Q_{30,10}}{s_3-s_1} = -6\sqrt{7}\,\Sigma_4 p_1^{13/6},\end{aligned}\qquad(3.32)$$

and the effective dielectric constant is given by

$$\frac{\epsilon_2-\epsilon_e}{\epsilon_2}\equiv F(s)\cong\frac{|M^{(1)}|^2}{s-s^{(1)}}+\frac{|M^{(3)}|^2}{s-s^{(3)}}$$

$$=\frac{p_1-252\Sigma_4^2p_1{}^{13/3}}{s-\frac{1}{3}+\frac{1}{3}p_1+24\Sigma_4^2p_1^{10/3}}+\frac{252\Sigma_4^2p_1^{13/3}}{s-\frac{3}{7}+\frac{60}{7}\Sigma_6p_1^{7/3}-24\Sigma_4^2p_1^{10/3}}\,. \tag{3.33}$$

Note that this form satisfies both of the sum rules proved in Ref. 1 up to and including terms of order $p_1^{13/3}$.

In order to compare our result with the one obtained by Rayleigh, we expand our result to get a power series in p_1 for $3/F(s)$, and use (2.6) to write s in terms of h. Doing this, we find

$$\frac{3}{F(s)}=\frac{2+h}{1-h}+p_1-16\frac{1-h}{\frac{4}{3}+h}\Sigma_4^2p_1^{10/3}\,. \tag{3.34}$$

This should be compared with Eq. (64) of Ref. 6, noting that Rayleigh's coefficient S_4 is related to our Σ_4 by

$$\Sigma_4p_1^{5/3}=S_4(a_0/b_0)^5\,, \tag{3.35}$$

where b_0 is the cubic-lattice parameter. The comparison shows that the last term in Rayleigh's Eq. (64) contains an extra factor $\frac{2}{5}$ which we think is wrong. The comparison also enables us to get the numerical value of Σ_4 from Rayleigh's approximate evaluation of S_4, namely, $S_4\cong 3.11$. In this way we find

$$\Sigma_4=(3/4\pi)^{5/3}S_4\cong 0.286\,. \tag{3.36}$$

Clearly, our expression (3.33) will give a better approximation to $F(s)$ in the vicinity of the leading poles than will Rayleigh's corrected expression (3.34). Thus, we can predict that for $p_1=0.2$, the second resonance will appear in optical experiments with a weight of about 0.019, while the first resonance will appear with a weight of about 0.18. Indications of such additional structure in the optical properties of a cubic array of Drude metal spheres have also been found recently by a numerical solution of Rayleigh's equations for the dielectric constant.[8] In a forthcoming publication we will examine this problem in detail and present results of numerical solutions of the eigenvalue equations for the poles and weights of cubic arrays of spherical inclusions with various values of p_1.[9]

In a recent series of articles, McKenzie and McPhedran[10] have pursued Rayleigh's approach to this problem by solving a truncated portion of his infinite set of equations. From a physical point of view, their truncation procedure involves ignoring higher electrostatic multipole moments which are induced in the spherical inclusions. In our procedure, we would also have to ignore higher multipole moments in order to be left with a finite matrix to be diagonalized. Our procedure encounters no difficulties near a pole of ϵ_e. In fact, because we calculate these poles directly, our results may even become more accurate in the vicinity of such a pole. On the other hand, the procedure of McKenzie and McPhedran, in which ϵ_e is calculated directly, would blow up near a pole of ϵ_e. We would therefore expect it to be of limited usefulness for calculating the optical properties of metal-insulator composites, where $\epsilon_1/\epsilon_2<0$. Indeed, it blows up even for $\epsilon_1>0$, $\epsilon_2=0$ when the spheres are nearly touching,[10] whereas we seem to encounter no serious diffulties at that point.[9]

IV. DISCUSSION

The scheme developed in this article offers a systematic way of getting approximations for the complex dielectric constant of a composite material whose microscopic geometry is known precisely. The scheme is strongly reminiscent of the tight-binding method for calculating the energy levels of a solid. It should be especially useful in cases where the composite is in the form of a periodic array of identical inclusions embedded in a homogeneous host. In that case, once the eigenstates of an individual grain are known, it should be possible to get very accurate results for the dielectric constant of the periodic composite by a numerical solution of the matrix eigenvalue problem of (3.22), properly truncated of course. Good results should be obtainable even for a volume fraction of the inclusions p_1 which is close to 1.

The significant general result for such systems is that the pole spectrum is discrete. This would be reflected in the optical properties of a composite made of metallic grains embedded in an insulating host. In the range of frequencies characterized by

$$1/\tau\ll\omega\ll\omega_p\,, \tag{4.1}$$

where τ is the relaxation time for dc conductivity in the metal while ω_p is the electronic plasma frequency of the metal, the dielectric constant of the metal is real and negative, and it can be quite large. At these frequencies, the dielectric constant of the insulating host ϵ_I is usually quite independent of ω, while that of the metal ϵ_M varies rather strongly. Typically, this variation is described by

$$\epsilon_M(\omega)\cong 1-\omega_p^2/\omega^2\,. \tag{4.2}$$

Thus, by varying ω, the ratio

$$s=\epsilon_I/(\epsilon_I-\epsilon_M) \tag{4.3}$$

can be made to approach the poles of $F(s)$, leading to resonances in the optical properties of the composite.

Such a resonance has indeed been observed experimentally.[11-13] Previous attempts to interpret this resonance by means of the Clausius-Mossotti approximation have met with a mixed success. Our discussion offers the chance for a more systematic approach to the interpretation of these observations.

What still needs to be done is to find a way to introduce a certain amount of disorder into the periodic array that we have been discussing. Presumably, if the disorder is not too great, the discrete resonances will only be slightly broadened, but will retain their identity. For a strong disorder, the smearing may become so pronounced that the resonance structure will disappear altogether. Indeed, for granular metal-insulator composites where a great deal of clumping of the metallic particles occurs, sharp resonances have not been observed in the optical properties.[14]

Finally, we predict that if the disorder is small, additional resonances will be observed in the optical behavior, such as the one whose properties were worked out in Sec. III for a simple cubic array of spheres.

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APPENDIX: THE MATRIX ELEMENT OF $\hat{G}$ BETWEEN EIGENSTATES OF TWO ISOLATED SPHERES

We wish to calculate the matrix element (3.13) between the lm eigenstate in a sphere of radius a_0 centered at the origin, and an $l'm'$ eigenstate in a sphere of radius a_1 centered at $\vec{b}$. The two spheres do not overlap. In order to evaluate the integral in (3.13), we shall have to expand the $l'm'$ function

$$\psi_{bl'm'}(\vec{\mathrm{r}})=\frac{a_1^{2l'+1}}{|\vec{\mathrm{r}}-\vec{\mathrm{b}}|^{l'+1}}Y_{l'm'}(\Omega_{r-b}), \quad \text{for } |\vec{\mathrm{r}}-\vec{\mathrm{b}}|>a_1, \tag{A1}$$

in spherical harmonics centered around the origin. This can be done with the help of Ref. 15, where explicit formulas are developed for the Helmholtz multipole matrix elements of the translation operator. From Eqs. (24), (13), and (19′) of that reference we can deduce the following equation, which expands the singular multipole field $n_{l'}(k|\vec{\mathrm{r}}-\vec{\mathrm{b}}|)Y_{l'm'}(\Omega_{r-b})$ as a sum of products of a regular multipole field $j_\lambda(kr)Y_{\lambda\mu}(\Omega)$ and a singular multipole field $n_L(kb)Y_{LM}(\Omega_b)$,

$$n_{l'}(k|\vec{\mathrm{r}}-\vec{\mathrm{b}}|)Y_{l'm'}(\Omega_{r-b})=\sum_{\substack{LM\\ \lambda\mu}} i^{L+\lambda-l'}(-1)^{L+m'}[4\pi(2L+1)(2l+1)(2\lambda+1)]^{1/2} \times\begin{pmatrix}L & l' & \lambda\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix}L & l' & \lambda\\ M & -m' & \mu\end{pmatrix}[j_\lambda(kr)Y_{\lambda\mu}(\Omega)][n_L(kb)Y_{LM}(\Omega_b)]. \tag{A2}$$

Here j_l and n_l are the regular and the singular spherical Bessel functions, and k is a wave-number which should be sent to zero in order to reduce these multipole solutions of the Helmholtz equation to the desired multipole solutions of Laplace's equation. When this is done, and all the powers of k are brought to the right-hand side, the appearance of the combination

$$\begin{pmatrix}L & l' & \lambda\\ 0 & 0 & 0\end{pmatrix}k^{l'+\lambda-L} \tag{A3}$$

ensures that, in the limit $k\to 0$, only the terms with $L=l'+\lambda$ survive in the sum. The orthogonality relation (3.4) can now be used to calculate the scalar product of this sum with ψ_{0lm}, which is given by

$$\psi_{0lm}(\vec{\mathrm{r}})=r^l Y_{lm}(\Omega), \quad \text{for } r<a_0. \tag{A4}$$

When the norms $\|\psi_{0lm}\|$, $\|\psi_{bl'm'}\|$ and the eigenvalue $s_{bl'm'}$ are properly included from (3.3) and (3.4), one obtains for the matrix element the expression of (3.28). In order to get that result we used the explicit expression for $3-j$ coefficients of the form[7]

$$\begin{pmatrix}l & l' & l+l'\\ m & m' & -m-m'\end{pmatrix}, \tag{A5}$$

as well as the connection between Y_{lm} and the associated Legendre polynomials $P_l^{(m)}$.

A more elementary derivation of the results of this Appendix is possible if instead of taking the $k \to 0$ limit in Helmholtz multipole fields, one uses the simpler electrostatic multipole fields from the outset. We will present such a derivation in a forthcoming publication.[9]

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[1] D. J. Bergman, Phys. Rep. 43, 377 (1978).

[2] This equation is the famous reciprocity principle of electrostatics. For a proof and a discussion, see, e.g., P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953), Vol. I, p. 808.

[3] R. Englman and R. Ruppin, J. Phys. C 1, 614–629 (1968); R. Ruppin and R. Englman, Rep. Prog. Phys. 33, 149–196 (1970).

[4] P. Clippe, R. Evrard, and A. A. Lucas, Phys. Rev. B 14, 1715–1721 (1976).

[5] R. Fuchs, AIP Conf. Proc. 40, 276 (1978).

[6] J. W. S. Rayleigh, Philos. Mag. 34, 481–502 (1892).

[7] The definition of $P_l^{(m)}$ and Y_{lm}, as well as the formulas for the 3-j coefficients which are needed in the Appendix, were taken out of A. Messiah, *Quantum Mechanics* (North-Holland, Amsterdam, 1962), pp. 492–496 and 1054–1060.

[8] W. T. Doyle, AIP Conf. Proc. 40, 300 (1978).

[9] J. Bar-Sagi and D. J. Bergman (unpublished).

[10] D. R. McKenzie and R. C. McPhedran, Nature 265, 128–129 (1977); R. C. McPhedran and D. R. McKenzie, Proc. R. Soc. London A 359, 45–63 (1977).

[11] R. W. Cohen, G. D. Cody, M. D. Coutts, and B. Abeles, Phys. Rev. B 8, 3689–3701 (1973).

[12] J. I. Gittleman and B. Abeles, Phys. Rev. B 15, 3273–3275 (1977).

[13] C. G. Granqvist and O. Hunderi, Phys. Rev. B 16, 3513–3534 (1977); Phys. Rev. B 16, 1353–1358 (1977).

[14] D. B. Tanner, A. J. Sievers, and R. A. Buhrman, Phys. Rev. B 11, 1330–1341 (1975).

[15] M. Danos and L. C. Maximon, J. Math. Phys. 6, 766–778 (1965).