Strongly nonlinear composite dielectrics: A perturbation method for finding the potential field and bulk effective properties

Raphael Blumenfeld
Cambridge University, The Cavendish Laboratory, Madingley Road, Cambridge CB3 0HE, United Kingdom

David J. Bergman
Raymond and Beverly Sackler Faculty of Exact Sciences, School of Physics and Astronomy, Tel-Aviv University, Tel-Aviv 69978, Israel
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A class of strongly nonlinear composite dielectrics is studied. We develop a general method to reduce the scalar-potential-field problem to the solution of a set of linear Poisson-type equations in rescaled coordinates. The method is applicable for a large variety of nonlinear materials. For a power-law relation between the displacement and the electric fields, it is used to solve explicitly for the value of the bulk effective dielectric constant ϵ_e to second order in the fluctuations of its local value. A simlar procedure for the vector potential, whose curl is the displacement field, yields a quantity analogous to the inverse dielectric constant in linear dielectrics. The bulk effective dielectric constant is given by a set of linear integral expressions in the rescaled coordinates and exact bounds for it are derived.

I. INTRODUCTION

A strongly nonlinear dielectric is one where the nonlinearity appears as the leading form of the behavior under the application of an external field, rather than as a small correction to a predominant linear response. Such a nonlinear behavior is quite common when a sufficiently strong field is applied to any condensed medium. But under static conditions it is usually difficult to apply the strong field necessary to reach the nonlinear regime of a dielectric. Nevertheless, there are special cases, where such a regime is attained without difficulty in other types of macroscopic response. For example, nonlinear electrical conductivity is found in many ceramic conductors,[1] while nonlinear elastic behavior is usually observed under stresses that approach breakdown threshold for rupture.[2] In dielectrics, nonlinear behavior is routinely observed under the intense ac fields that are available from lasers. Our motivation for the present study is mainly theoretical: nonlinear response is notoriously difficult to handle. Except when the medium as well as the boundary conditions are completely uniform, there are almost no known methods for calculating the behavior of such systems, other than by brute force numerical solution of the appropriate nonlinear differential equation.

In this paper we study the problem of calculating the bulk effective macroscopic properties of a strongly nonlinear composite medium. Specifically, we address the case of a nonlinear dielectric composite, whose components are isotropic and obey a local constitutive relation of the form

$$\mathbf{D}(\mathbf{r})=\epsilon(\mathbf{r})|E(\mathbf{r})|^{\beta}\mathbf{E}(\mathbf{r}), \tag{1.1}$$

where the nonlinearity exponent β is the same everywhere, but the nonlinear dielectric constant $\epsilon(\mathbf{r})$ differs from component to component. The macroscopic behavior of the medium is described by the relation between the volume averaged electric field $\langle \mathbf{E} \rangle$ and the volume averaged displacement $\langle \mathbf{D} \rangle$. It can be shown that under the above assumptions, and when the composite is isotropic, this relation has the form

$$\langle \mathbf{D} \rangle=\epsilon_e|\langle E \rangle|^{\beta}\langle \mathbf{E} \rangle ,$$

where ϵ_e is the bulk effective nonlinear constant, which, like $\epsilon(\mathbf{r})$, is independent of the field $\mathbf{E}(\mathbf{r})$. To calculate ϵ_e one needs to know the local field within the medium under boundary conditions that would result in a uniform field $\mathbf{E}(\mathbf{r})\equiv\mathbf{E}_0$ if the medium were homogeneous. Our approach to solving this problem is to expand $\mathbf{E}(\mathbf{r})\equiv-\nabla\Phi(\mathbf{r})$, and consequently ϵ_e, in powers of the spread $\delta\epsilon$ of the different possible dielectric constants of the components.

Our main results are the following.

(1) The electric potential field, $\Phi(\mathbf{r})$ can be found, to any finite order in $\delta\epsilon$, by solving a hierarchy of *linear Poisson equations* in the composite medium.

(2) The bulk effective nonlinear dielectric constant ϵ_e is expressed, to any finite order in $\delta\epsilon$, as an explicit integral involving $\Phi(\mathbf{r})$ up to a lower order in $\delta\epsilon$.

(3) The above two results are actually applicable to a more general class of nonlinear constitutive relations than that expressed by (1.1).

(4) The value of ϵ_e is found *exactly* to second order in $\delta\epsilon$, extending a classic result for the linear case.[3] An analogous treatment yields the corresponding value of the quantity $\omega\equiv\epsilon^{-1/(\beta+1)}$. To this order, both results are found to be *independent* of the detailed microgeometry of the composite.

(5) Exact bounds are found for the value of ϵ_e.

The outline of the rest of this paper is as follows: In Sec. II we expand the scalar potential field $\Phi(\mathbf{r})$ around its value in a homogeneous medium subject to the same boundary conditions. We show that each term in this expansion can be found by solving a linear differential equa-

tion of the Poisson type. In Sec. III we present an expression for the nth order term in the expansion of ϵ_e by using the solution for Φ only up to order $(n-2)$. For completeness, we give the exact result for ϵ_e to second order in $\delta\epsilon$.[4] An analogous result for ω_e (defined above) is also presented, but the derivation is given in the Appendix.

II. EXACT ITERATIVE PROCEDURE FOR THE SOLUTION OF THE SCALAR ELECTROSTATIC POTENTIAL FIELD

Consider a charge-neutral medium composed of grains, each of which is assumed to be homogeneous in its dielectric properties. Different grains are assumed to have different dielectric constants ϵ_i. Each of the grains follows (1.1) with the value of β being identical for all of them. The system occupies a volume between two conducting plates at $z=0$ and at $z=L$, and is assumed to be very large in all other dimensions. The potentials on the plates are $\Phi(z=0)=0$ and $\Phi(z=L)=E_0L$. Altogether the material is composed of N different components with ϵ_i $(i=1,2,\ldots,N)$, distributed in the system with respective concentrations p_i. The local dielectric constant can be written as

$$\epsilon(\mathbf{r})=\sum_i \epsilon_i \Theta_i(\mathbf{r}) , \tag{2.1}$$

where Θ_i is the step function whose value is 1 within the grains of the ith component and 0 elsewhere. The volume average of the local dielectric constant is

$$\langle \epsilon \rangle = \frac{1}{V}\int \epsilon(\mathbf{r}) d^d r = \sum_i p_i \epsilon_i . \tag{2.2}$$

Under the above imposed boundary conditions, a homogeneous system having a dielectric constant ϵ_0 gives rise to a constant field in the z direction, $\mathbf{E}_0$. The value of ϵ_0, around which ϵ_e (and everything else) is expanded, can be arbitrary.

We proceed to show that the potential field can be generally found via an iterative procedure. Before embarking upon this calculation, it should be noted that there are cases where an inhomogeneous medium can be solved exactly in a straightforward manner. These cases are special due to the particular geometrical internal structure of the composite. One such case consists of regions of different dielectric constants whose interfaces are parallel to the field and perpendicular to the capacitor plates [Fig. 1(a)]. In this configuration $\epsilon(\mathbf{r})$ is constant along any path in the direction of the field. Since the tangential component of $\mathbf{E}$ must be continuous at the interfaces, the internal field remains constant and perpendicular to the plates, i.e., $\mathbf{E}=E_0\hat{\mathbf{z}}$ everywhere. Consequently, the displacement field $\mathbf{D}$ varies in a simple way within this composite. Thus, using (2.1), the volume average of $\mathbf{D}$ is found to be

$$\langle D \rangle = \frac{1}{V}\int \epsilon(\mathbf{r}) E_0^{\beta+1} d^d r = E_0^{\beta+1}\sum_i p_i \epsilon_i , \tag{2.3}$$

where the summation is over the i different values of the dielectric constant. It follows that

$$\epsilon_e = \sum_i p_i \epsilon_i . \tag{2.4}$$

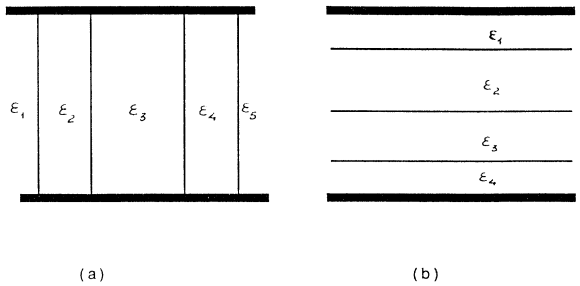

FIG. 1. An inhomogeneous nonlinear dielectric system with regions of different values of e that are arranged (a) as parallel (not necessarily circular) cylinders with interfaces that are perpendicular to the capacitor plates and (b) as flat slabs that are parallel to the capacitor plates.

Another solvable geometry consists of slabs of regions with different dielectric constants that lie in parallel to the plates [Fig. 1(b)]. On an interface between any two regions the normal component of $\mathbf{D}$ must be continuous approaching from both sides. Within each region the medium is homogeneous so that $\mathbf{D}$ does not vary. It follows that $\mathbf{D}$ does not change along a path perpendicular to the plates and $\mathbf{D}=D_0\hat{\mathbf{z}}$ is constant over the entire volume of the system. Averaging the electric field over the volume yields

$$\langle E \rangle = \frac{1}{V}\int \left[\frac{D_0}{\epsilon}\right]^{1/(\beta+1)} d^d r = D_0^{1/(\beta+1)}\sum_i p_i \epsilon_i^{1/(\beta+1)} . \tag{2.5}$$

Hence the bulk effective dielectric constant is

$$\epsilon_e = -\left[\sum_i p_i \epsilon_i^{-1/(\beta+1)}\right]^{-(\beta+1)} . \tag{2.6}$$

These two microgeometries correspond to nonlinear conductors in parallel and in series, respectively, and indeed the forms of ϵ_e resemble very much the corresponding effective conductances of those configurations.[5-7]

We now turn to discuss general composites. In a homogeneous medium the field is uniform $E_0\hat{\mathbf{z}}=-\nabla\Phi_0$, and hence we expect that if the local dielectric constant can be expanded around some constant value ϵ_0, the potential Φ can be also expanded around Φ_0, i.e., $\Phi=\Phi_0+\sum_{n=1}^{\infty}\delta^n\Phi$. To implement this we first write (1.1) in a more general form

$$\mathbf{D}=\epsilon(\mathbf{r}) f(E^2)\mathbf{E} , \tag{2.7}$$

and consider its nth variation

$$\delta^n\mathbf{D}/\epsilon_0 = f(E_0^2)\delta^n\mathbf{E} + 2f'(E_0^2)(\mathbf{E}_0\cdot\delta^n\mathbf{E})E_0 - F_0(\{\delta^m\mathbf{E}\}; m<n) , \tag{2.8}$$

where F_0 is some function that contains correction terms

to $\mathbf{E}$ of order strictly less than n, along with powers of $\delta\epsilon \equiv \epsilon(\mathbf{r}) - \epsilon_0$. Expanding the equation

$$\mathrm{div}\mathbf{D}(\mathbf{r}) = 0 \tag{2.9}$$

term by term, we obtain the following form for $\delta^n\Phi$,

$$\left[\nabla^2 + 2\left[\frac{\partial \ln f(x)}{\partial \ln x}\right]_{x=E_0^2}\partial_{zz}\right]\delta^n\Phi = F_1 , \tag{2.10}$$

where F_1 depends only on terms in the expansion of Φ that are of order strictly lower than n. The left-hand side (lhs) of (2.10) can be now transformed by rescaling the coordinates as follows:

$$x \to x, \quad y \to y, \quad z \to \zeta = z/\sqrt{B+1} ,$$

where

$$B = 2\left[\frac{\partial \ln f(x)}{\partial \ln x}\right]_{x=E_0^2} .$$

This transformation yields, in the rescaled coordinates,

$$\nabla^2\delta^n\Phi = F_2(\epsilon_0, \delta\epsilon, E_0, \delta E, \ldots, \delta^{n-1}E) , \tag{2.11}$$

which has the form of a *linear Poisson equation.* It is true that for each set of boundary conditions the z axis is rescaled differently, but this should not pose a problem, since the scaling factor B depends only on the average field $\mathbf{E}_0$, which is independent of the spatial coordinates. Equations of this type have been studied extensively, and a large body of knowledge exists concerning approaches for solving them. Thus, in principle, (2.11) provides a systematic iterative procedure for calculating the nth order correction to the potential Φ_0 that uses solutions for lower-order corrections.

In the particular case of power-law nonlinearity as in (1.1), $B = \beta$. It is easy to see that only in this case, is B independent of E_0. Thus we obtain the following equation for the first variation $\delta\Phi$

$$\nabla^2\delta\Phi = \frac{1}{\sqrt{\beta+1}}\mathrm{div}(\delta\eta\mathbf{E}_0) , \tag{2.12}$$

where $\eta \equiv \epsilon/\epsilon_0$ and $\delta\eta = \eta - 1$. A formal solution to (2.12) can be written as

$$\delta\Phi(\boldsymbol{\rho}) = -\frac{E_0}{\sqrt{\beta+1}}\int G(\boldsymbol{\rho},\boldsymbol{\rho}')\partial_{\zeta'}\delta\eta(\boldsymbol{\rho}')d^d\rho' , \tag{2.13}$$

in terms of a Green's function $G(\boldsymbol{\rho},\boldsymbol{\rho}')$, that satisfies

$$\nabla^2 G = -\delta^d(\boldsymbol{\rho} - \boldsymbol{\rho}') ,$$

$G = 0$ on the boundaries. Here the integration is carried out in the rescaled coordinates $\boldsymbol{\rho}'$. Integrating (2.13) by parts and using the condition that $G = 0$ on the boundary, it is found that

$$\delta\Phi(\boldsymbol{\rho}) = -\frac{E_0}{\sqrt{\beta+1}}\int \partial_{\zeta'}G(\boldsymbol{\rho},\boldsymbol{\rho}')[\delta\eta(\boldsymbol{\rho}') - \langle\delta\eta\rangle]d^d\rho' , \tag{2.14}$$

where the inclusion of the constant $\langle\delta\eta\rangle$ in the integrand contributes nothing to the integral due to the zero boundary condition on G. The purpose of this inclusion will become apparent below. The variables $\boldsymbol{\rho}$ and $\boldsymbol{\rho}'$ are position vectors in the rescaled coordinates. A similar procedure yields the following formal solution for the second correction to Φ

$$\delta^2\Phi(\boldsymbol{\rho}) = \frac{2\beta E_0}{\sqrt{\beta+1}}\delta\Phi\partial_\zeta\delta\Phi + \int d^d\rho'\left[\frac{\beta E_0}{\sqrt{\beta+1}}\partial_{\zeta'}\delta\Phi - \delta\eta\right]\nabla'\delta\Phi\cdot\nabla' G(\boldsymbol{\rho},\boldsymbol{\rho}')$$
$$+\frac{\beta E_0}{\sqrt{\beta+1}}\int d^d\rho' G(\boldsymbol{\rho},\boldsymbol{\rho}')\partial_{\zeta'}[\delta\Phi(\nabla')^2\delta\Phi + 2(\partial_{\zeta'}\delta\Phi)^2] . \tag{2.15}$$

Substituting (2.14) into (2.15) gives a combination of double integrals containing products of the form $\delta\eta(\boldsymbol{\rho})\delta\eta(\boldsymbol{\rho}')$. In general, to solve for the nth order of Φ one has to consider n-tuple integrals over $\boldsymbol{\rho}_1, \boldsymbol{\rho}_2, \ldots, \boldsymbol{\rho}_n$. These integrals contain combinations of derivatives of Green's functions $G(\boldsymbol{\rho},\boldsymbol{\rho}_1), G(\boldsymbol{\rho}_1,\boldsymbol{\rho}_2)$, etc., with products of the form $\delta\eta(\boldsymbol{\rho}_1)\delta\eta(\boldsymbol{\rho}_2)\cdots\delta\eta(\boldsymbol{\rho}_n)$. This multiple product can be expressed in terms of products of differences of the form $\delta\eta(\boldsymbol{\rho}_i) - \langle\delta\eta\rangle$, that are mathematically well behaved (namely, the volume average of each of them vanishes). When the composite is macroscopically uniform, averages of such products depend only on relative coordinates. It can then be shown that when the system is large compared to the lengths of spatial decay of these averages, we can replace the above products in the calculation of ϵ_e by their averages. This is demonstrated below in the calculation of $\delta^2\epsilon_e$. Thus our method enables to reduce the calculation of ϵ_e of the nonlinear medium to a set of weighted n-point correlation functions of $\delta\epsilon(\boldsymbol{\rho})$.

III. CALCULATION OF ϵ_e TO SECOND ORDER IN $\delta\epsilon$

We next proceed to find the first and second-order corrections to ϵ_e, $\delta\epsilon_e$, and $\delta^2\epsilon_e$, exactly. Starting from the energy density[4]

$$u = \frac{\beta+1}{4\pi(\beta+2)V}\int \mathbf{E}\cdot\mathbf{D}\, d^d r \tag{3.1}$$

and carrying out a first variation calculation on both sides yields

$$\delta\epsilon_e = \frac{1}{V}\int\left[\delta\epsilon(\mathbf{r})\left[\frac{|\nabla\Phi|}{E_0}\right]^{\beta+2} + E_0^{-(\beta+2)}\epsilon(\mathbf{r})\delta(|\nabla\Phi|^{\beta+2})\right]d^d r . \tag{3.2}$$

The second term on the right-hand side (rhs) can be expanded and integrated by parts to yield

$$\frac{\beta+2}{E_0^{\beta+2}}\left[\oint_S \delta\Phi\epsilon(\mathbf{r})|\nabla\Phi|^{\beta}\nabla\Phi\cdot d\mathbf{S}-\frac{1}{V}\int \delta\Phi\,\mathrm{div}\mathbf{D}\,d^d r\right].$$

Both terms within the rectangular brackets vanish—the first, since $\delta\Phi=0$ on the boundary, and the second because of (2.9). This leaves

$$\delta\epsilon_e=\frac{1}{V}\int \delta\epsilon(\mathbf{r})\left[\frac{|\nabla\Phi|}{E_0}\right]^{\beta+2} d^d r\ . \tag{3.3}$$

Therefore, to first order, one can replace Φ by Φ_0 to get

$$\delta\epsilon_e=\langle\epsilon\rangle-\epsilon_0\ . \tag{3.4}$$

This coincides with the exact result (2.4) for the real system shown in Fig. 1(a). It is now easy to show that $\epsilon_e\le\langle\epsilon\rangle$, as in linear composites.[8,9] Since (3.1) expresses the total energy (divided by the volume) in the system, which is minimal, any trial function for $\mathbf{E}$ that satisfies the boundary conditions yields a larger value for ϵ_e when substituted for the actual field in the integrand. In particular, choosing $\mathbf{E}_0$ as the trial function, one immediately verifies the above claim. Moreover, since $\epsilon_e=\langle\epsilon\rangle$ comprises a *realizable* upper bound on ϵ_e (see above), then this bound is optimal under the given information on the system. From (3.4) one can also conclude that ϵ_0 cancels out to first order.

Applying the δ operator again to relation (3.3) we obtain the second variation

$$\delta^2\epsilon_e=\frac{(\beta+2)E_0^{-(\beta+2)}}{V}\int \delta\epsilon(\mathbf{r})|\nabla\Phi|^{\beta}(\nabla\Phi\cdot\nabla\delta\Phi)d^d r\ . \tag{3.5}$$

In general, the nth order correction to ϵ_e is

$$\delta^n\epsilon_e=\frac{E_0^{-(\beta+2)}}{V}\int \delta\epsilon(\mathbf{r})\delta^{n-1}(|\nabla\Phi|^{\beta+2})d^d r\ . \tag{3.6}$$

As can be seen from (3.5), the nth order correction to ϵ_e requires the knowledge of Φ up to, at most, order $(n-1)$. As it turns out, for all $n\ge 3$, one actually needs to know Φ only up to order $n-2$ as we now proceed to show. We first demonstrate explicitly that $\delta^3\epsilon_e$ depends only on $\delta\Phi$. Starting from (3.5), integrating by parts and discarding the surface term, one obtains

$$\delta^2\epsilon_e=-\frac{(\beta+2)E_0^{-(\beta+2)}}{V}\times\int \delta\Phi\,\mathrm{div}[\delta\epsilon|\nabla\Phi|^{\beta}\nabla\Phi]d^d r\ . \tag{3.7}$$

From (2.9) the following equality can be extracted:

$$\mathrm{div}[\epsilon\,\delta^n(|\nabla\Phi|^{\beta}\nabla\Phi)]=-n\,\mathrm{div}[\delta\epsilon\,\delta^{n-1}(|\nabla\Phi|^{\beta}\nabla\Phi)]\ . \tag{3.8}$$

Substituting (3.8) with $n=1$ into (3.7) and retracing the integration by parts we obtain

$$\delta^2\epsilon_e=-\frac{(\beta+2)E_0^{-(\beta+2)}}{V}\int \epsilon\,\delta(|\nabla\Phi|^{\beta}\nabla\Phi)\cdot\nabla\delta\Phi\,d^d r\ . \tag{3.9}$$

Applying now a small variation to both sides of (3.9) and using (3.8) with $n=2$ one gets

$$\delta^3\epsilon_e=-\frac{(\beta+2)E_0^{-(\beta+2)}}{V}\left[-\int \delta\epsilon\,\delta(|\nabla\Phi|^{\beta}\nabla\Phi)\cdot\nabla\delta\Phi\,d^d r+\int \epsilon\,\delta(|\nabla\Phi|^{\beta}\nabla\Phi)\cdot\nabla\delta^2\Phi\,d^d r\right]. \tag{3.10}$$

It can be shown that the following is an identity:

$$\delta(|\nabla\Phi|^{\beta}\nabla\Phi)\cdot\nabla\delta^2\Phi=\delta^2(|\nabla\Phi|^{\beta}\nabla\Phi)\cdot\nabla\delta\Phi-\beta|\nabla\Phi|^{\beta-4}(\nabla\phi\cdot\nabla\delta\Phi)[(\beta-2)(\nabla\Phi\cdot\nabla\delta\Phi)^2+3|\nabla\Phi|^2\nabla\delta\Phi|^2]\ . \tag{3.11}$$

Substituting this into (3.10) and using (3.8) for the integration of the first term on the rhs of (3.11), yields finally

$$\delta^3\epsilon_e=\frac{(\beta+2)E_0^{-(\beta+2)}}{V}\left[3\int \delta\epsilon\,\delta(|\nabla\Phi|^{\beta}\nabla\Phi)\cdot\nabla\delta\Phi\,d^d r+\beta\int \epsilon|\nabla\Phi|^{\beta-4}(\nabla\Phi\cdot\nabla\delta\Phi)[(\beta-2)(\nabla\Phi\cdot\nabla\delta\Phi)^2+3|\nabla\Phi|^2|\nabla\delta\Phi|^2]d^d r\right]. \tag{3.12}$$

Thus the third-order correction to ϵ_e depends only on Φ and $\delta\Phi$. Further application of the δ operator $n-3$ times to (3.12) yields that the nth-order correction to ϵ_e depends on Φ only up to order $n-2$ as claimed above.

We now proceed to calculate the bulk effective dielectric constant explicitly to second order. Note that in the linear case this calculation is done under the following assumptions: (i) the correlation between fluctuations of $\epsilon(\mathbf{r})$ at different points is short ranged compared to the size of the system, (ii) the material is homogeneous on length scales that are much larger than the correlation length, and (iii) the medium is isotropic. We now extend this calculation to the nonlinear case with *no additional assumptions.*

Using $\delta\Phi=0$ on the boundary, one can integrate (3.5) by parts and observe that adding a constant to $\delta\epsilon(\mathbf{r})$ leaves the result unchanged. In particular, it is possible to replace $\delta\epsilon(\mathbf{r})$ in (3.5) by $[\delta\epsilon(\mathbf{r})-\langle\delta\epsilon\rangle]$. Substituting relation (2.13) for $\delta\Phi$ in (3.5) and changing the integration variables to the rescaled coordinates yields

$$\delta^2\epsilon_e=\frac{\epsilon_0(\beta+2)}{\sqrt{\beta+1}V}\int d^d\rho\int d^d\rho'\{[\delta\eta(\boldsymbol{\rho})-\langle\delta\eta\rangle][\delta\eta(\boldsymbol{\rho}')-\langle\delta\eta\rangle]\}\partial_{\xi\xi'}G(\boldsymbol{\rho},\boldsymbol{\rho}')\ . \tag{3.13}$$

Changing variables to ρ and to $\mathbf{R}=\rho'-\rho$ and integrating over ρ first, at fixed $\mathbf{R}$, one can see that for large values of $\mathbf{R}$ (above the inhomogeneity correlation length) the integral vanishes due to the independence of the fluctuations in $\eta(\rho)$ and $\eta(\rho+\mathbf{R})$. For small values of $\mathbf{R}$ we can use the Green's function, given below, which depends only on $\mathbf{R}$, and identify the integral over ρ as the average of the term in the curly braces. Therefore, we can replace this term by the correlation function $g(\rho,\rho')=\langle[\delta\eta(\rho)-\langle\delta\eta\rangle][\delta\eta(\rho')-\langle\delta\eta\rangle]\rangle$ to obtain

$$\delta^2\epsilon_e=\frac{\epsilon_0(\beta+2)}{\sqrt{\beta+1}V}\int\int g(\rho,\rho')\partial_{\xi\xi'}G(\rho,\rho')d^d\rho\, d^d\rho' . \quad (3.14)$$

If the composite is macroscopically homogeneous, then g depends only on the relative coordinate $\mathbf{R}$. Assuming further than $g(\mathbf{R})$ decays to zero over a distance much smaller than the size of the system, one can replace G over most of the volume (except near the boundaries) by

$$G_0(\mathbf{R})=1/(4\pi|\mathbf{R}|) . \quad (3.15)$$

One of the volume integrations now becomes trivial and yields the rescaled volume, $\Omega=V/\sqrt{\beta+1}$, leaving, for $d>2$,

$$\delta^2\epsilon_e=-\frac{\epsilon_0(\beta+2)}{\beta+1}\int d^dR\, g(\mathbf{R})\partial_{\xi\xi'}G_0(\mathbf{R}) . \quad (3.16)$$

It should be emphasized that this relation depends only on the assumed macroscopic homogeneity and the short range of correlations between the values of $\delta\epsilon(\mathbf{r})$ at different locations. We now invoke the assumption of isotropy, namely, that $g(\mathbf{R})$ is spherically symmetric in the *original* coordinates. Since (3.16) is carried out in the *rescaled* coordinates, the correlation function g has a spheroidal symmetry, i.e., it is stretched or contracted in the ζ direction, depending on the sign of β. For $\beta=0$ (the linear case) the integral can be evaluated exactly by using the spherical symmetry, leading to a result that depends only on $g(0)$.[8] We now show that (3.16) can also be evaluated in the present case. Consider the Fourier transforms of the Green's function, $\tilde{G}_0=1/|k|^2$, and of g in the rescaled coordinates

$$\tilde{g}=\int g(\rho)e^{i\mathbf{k}\cdot\rho}d^d\rho=\frac{1}{\sqrt{\beta+1}}\int g_0(\mathbf{R})\exp[i(\mathbf{k}_t\cdot\mathbf{x}_t+k_z z/\sqrt{\beta+1})]\,d^dR$$

$$=\frac{1}{\sqrt{\beta+1}}\tilde{g}_0(k_1,k_2,\ldots,k_z/\sqrt{\beta+1}) , \quad (3.17)$$

where in the second form we transformed the integral back to the original coordinate system. The variables $\mathbf{k}_t$ and $\mathbf{x}_t$, are, respectively, the components of $\mathbf{k}$ and $\mathbf{R}$ that are orthogonal to z, and g_0 is the spherically symmetric correlation function in the original coordinates. Since $\tilde{g}_0(\mathbf{q})$ depends only on q^2 one can use (3.17) to relate between the Fourier transforms in the different coordinate systems

$$\tilde{g}(\mathbf{k})=\frac{1}{\sqrt{\beta+1}}\tilde{g}_0\left[k^2\left[1-\frac{\beta}{\beta+1}\cos^2\theta\right]\right] , \quad (3.18)$$

where θ is the polar angle between $\mathbf{k}$ and the ζ axis. Substituting this expression and the Fourier transform of G_0 into (3.16) one obtains

$$\delta^2\epsilon_e=-\frac{(\beta+2)\epsilon_0}{(\beta+1)}\int d^dR\int\frac{d^dq}{(2\pi)^d}\int\frac{d^dk}{(2\pi)^d}\tilde{g}(\mathbf{k})\left[\frac{q_\zeta}{q}\right]^2 e^{-i\mathbf{R}\cdot(\mathbf{q}+\mathbf{k})} . \quad (3.19)$$

The integration over $\mathbf{R}$ and $\mathbf{q}$ can be carried out to yield

$$\delta^2\epsilon_e=\frac{(\beta+2)\epsilon_0}{\beta+1}\int\frac{d^dk}{(2\pi)^d}\tilde{g}(\mathbf{k})\cos^2\theta . \quad (3.20)$$

Rewriting this integral using polar coordinates in k-space, we change the radial coordinate from k to K defined by

$$K\equiv\left[1-\frac{\beta}{\beta+1}\cos^2\theta\right]^{1/2}k ,$$

and use (3.18) to substitute for $\tilde{g}(\mathbf{k})$. The azimuthal angle is integrated trivially, leaving integrals over K and the polar variable $u=\cos\theta$

$$\delta^2\epsilon_e=-\frac{(\beta+2)\epsilon_0}{2(\beta+1)^{3/2}}\int_0^\infty\tilde{g}_0(K^2)\frac{S_dK^{d-1}}{(2\pi)^d}dK\int_{-1}^{1}\frac{u^2}{[1-(\beta u^2/\beta+1)]^{d/2}}du , \quad (3.21)$$

where S_d is the surface area of a unit hypersphere in d dimensions [$S_d=2\pi$, 4π, and $2\pi^{d/2}/\Gamma(d/2)$ in 2, 3, and d dimensions, respectively). The integral over K can be identified as $g_0(0)$. By changing variables to $\sin x\equiv\sqrt{\beta/(\beta+1)}u$, the second integral becomes

$$2\left[\frac{\beta+1}{\beta}\right]^{3/2}\int_0^{\arcsin\sqrt{\beta/(\beta+1)}}(\cos^{1-d}x-\cos^{3-d}x)dx\ . \tag{3.22}$$

Calculating this integral in three dimensions and inserting into (3.21) finally yields

$$\delta^2\epsilon_e=-\frac{\epsilon_0(\beta+2)}{\beta}g_0(0)\left[1-\frac{1}{\sqrt{\beta}}\arcsin\left[\frac{\beta}{\beta+1}\right]^{1/2}\right]\ . \tag{3.23}$$

Note that this relation is valid for both $\beta>0$ and $-1<\beta\leq 0$ by analytic continuation, where the periodic function changes to a hyperbolic function, while $\delta^2\epsilon_e$ remains real. Recalling the definition of the correlation function $g_0(\mathbf{R})$, we have

$$g_0(0)=\langle[\delta\eta(\mathbf{r})-\langle\delta\eta\rangle]^2\rangle=\langle\eta^2\rangle-\langle\eta\rangle^2\ ,$$

and summing the zero- and first-order contributions, one finds[4]

$$\epsilon_e=\langle\epsilon\rangle-\frac{\beta+2}{2\epsilon_0\beta}\left[1-\frac{1}{\sqrt{\beta}}\arcsin\left[\frac{\beta}{\beta+1}\right]^{1/2}\right](\langle\epsilon^2\rangle-\langle\epsilon\rangle^2)+O((\delta\epsilon)^3)\ . \tag{3.24}$$

When $\beta\rightarrow 0$ this reduces to the well-known expression for the linear case,[3]

$$\epsilon_e(\beta=0)=\langle\epsilon\rangle-(\langle\epsilon\rangle^2-\langle\epsilon\rangle^2)/3\epsilon_0\ .$$

It is interesting to note that the result (3.24) is *independent* of the local microgeometry and only depends on the variance of the distribution of $\epsilon(\mathbf{r})$. Note also that, while (3.4) holds generally, the result for the second order (3.23) is valid only for an isotropic medium.

A procedure, similar to the above, can be carried out to find the bulk effective value of $\omega\equiv\epsilon^{-\gamma}[\gamma=1/(\beta+1)]$ to second order in $\delta\omega(r)$. From (2.9) it is clear that for a neutral medium one can define a vector potential $\mathbf{A}$ such that $\mathbf{D}=\nabla\times\mathbf{A}$. The basic relations that now define the mathematical problem are (analogously to the linear case[10]):

$$\mathbf{E}(\mathbf{r})=\omega(\mathbf{r})|\mathbf{D}(\mathbf{r})|^{\gamma-1}\mathbf{D}\ , \tag{3.25}$$

$$\nabla\times\mathbf{E}(\mathbf{r})=0\ , \tag{3.26}$$

and

$$\omega_e=\frac{D_0^{-(\gamma+1)}}{V}\int\omega(\mathbf{r})|\nabla\times\mathbf{A}|^{\gamma+1}d^dr\ . \tag{3.27}$$

As in the former case, the second-order correction to ω_e can be shown to depend only on the first-order correction to $\mathbf{A}$. The procedure in this case is somewhat more involved due to two complications: (i) To transform the differential equation for $\delta\mathbf{A}$ to the desired linear form it is not enough to rescale the coordinates, but rather, one also needs to rescale the magnitudes of the components of $\mathbf{A}$. (ii) A technical complication arises from the tensorial nature of the corresponding Green's function. Nevertheless, the calculation can be carried out explicitly (see the Appendix). The resulting expression that we find for ω_e is [in terms of β for comparison with (3.24)]

$$\omega_e=\langle\omega\rangle-\frac{\beta+2}{2\beta\omega_0}[\sqrt{\beta}(\beta+1)\arcsin\sqrt{\beta/(\beta+1)}-1](\langle\omega^2\rangle-\langle\omega\rangle^2)+O((\delta\omega)^3), \tag{3.28}$$

which agrees with the one found for ϵ_e to this order. This agreement is not surprising, since by their definitions $\epsilon_e=\omega_e^{-1/\gamma}$.[4] Hence an expansion of the quantities ϵ_e and $\omega_e^{-1/\gamma}$, in any variable, should agree to any other. As in the previous case, ω_e can be bounded by its volume average (see the discussion above)

$$\omega_e\leq\langle\omega\rangle=\sum_i p_i\epsilon_i^{-\gamma}\ . \tag{3.29}$$

This is the result of the variational property of (3.27): ω_e achieves its minimal value, under all possible vector fields $\mathbf{A}$ that satisfy the appropriate boundary conditions, only for the field that solves the appropriate differential equation. Thus, replacing the minimizing displacement field by a constant field $\mathbf{D}_0$ yields the above result. The rhs of (3.29) is realized for the real system shown in Fig. 1(b). Therefore, this bound is optimal for any medium that follows the above power-law nonlinear relation. This result, combined with the above bound on ϵ_e [see the discussion following (3.4)], yields the following bounds on ϵ_e

$$(\langle\epsilon^{-\gamma}\rangle)^{-1/\gamma}\leq\epsilon_e\leq\langle\epsilon\rangle\ . \tag{3.30}$$

These bounds are optimal given that the volume fractions of the components are the only information available about their spatial distribution.

IV. DISCUSSION AND CONCLUSIONS

In this paper we have discussed inhomogeneous, strongly nonlinear materials. The nonlinear equation for the scalar potential field (for nonlinear dielectrics) has been studied and a method to reduce it into a set of linear equations of the Poisson type, has been developed. Our method describes an operative procedure to solve iteratively for the scalar potential field, without having to cope directly with the nonlinear equation. The strongly nonlinear problem is reduced to a set of linear equations that are much easier to solve than the original problem. This procedure is formally applicable to any nonlinear dielectric whose local relation between $\mathbf{D}$ and $\mathbf{E}$ has the form $\mathbf{D}=\epsilon(\mathbf{r})f(E^2)\mathbf{E}$. The advantage of this method lies in the fact that there exists a large body of knowledge with regard to the solution of equations of the Poisson's type, e.g., conformal mapping, Green's functions, and also many numerical methods. We have used the method of the Green's function to calculate the bulk effective dielectric constant ϵ_e to second order in the spread of values of $\delta\epsilon$, assuming that the medium is isotropic and macroscopically homogeneous. The result is found to be independent of the detailed microgeometry, as in the linear case under similar assumptions.[8] Thus our calculation extends the classic result for linear composites.[3,8,9] When the local fluctuations in the dielectric constant are narrowly distributed [namely, $\delta\epsilon(\mathbf{r})\ll\langle\epsilon\rangle$], one can use this result to estimate the bulk dielectric constant with good accuracy. The higher-order corrections to ϵ_e have been cast in terms of integral expressions in the rescaled coordinates, with each order depending only on corrections of lower order to the local potential Φ. However, since even in the linear problem these expressions are very difficult to evaluate, the practical usefulness of this reduction for $n\geq 3$ may be limited. The problem lies in the need to know n-point correlation functions within the medium for increasingly large values of n, which is usually very difficult. Nevertheless, it should be noted that this difficulty does not stem from the nonlinearity of the medium but rather from its geometrical structure and is not very different from the difficulties encountered in linear dielectrics.

Although the above results have been obtained for dielectric systems, they apply to other problems, such as heat conductivity, electric conductivity, nonlinear magnetic behavior, etc. All these systems satisfy two equations of the type (1.1) and (2.9). While for the electric field $\mathbf{E}$ we treated in the text a scalar potential, due to $\nabla\times\mathbf{E}=\mathbf{0}$, one can analogously consider a vector potential field $\mathbf{A}$ whose curl is the displacement field $\mathbf{D}$, invert relation (1.1) to the form $\mathbf{E}(\mathbf{D})$, write $\nabla\times\omega(\nabla\times\mathbf{A})=\mathbf{0}$, and solve for $\mathbf{A}$ (see the Appendix). The resulting bulk effective value of ω_e agrees with ϵ_e in each order of δe, so that solving for $\mathbf{A}$ constitutes an alternative procedure for finding the effective bulk behavior.

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APPENDIX: THE CALCULATION OF ω_e TO SECOND ORDER IN THE FLUCTUATIONS

The nonlinear relation between $\mathbf{D}$ and $\mathbf{E}$ can be written in the form

$$\mathbf{E}(\mathbf{r})=\omega(\mathbf{r})|D(\mathbf{r})|^{\gamma-1}\mathbf{D}(\mathbf{r}),\quad \omega(\mathbf{r})\equiv[\epsilon(\mathbf{r})]^{-\gamma} \tag{A1}$$

where $\gamma\equiv 1/(\beta+1)$. This relation is complemented by

$$\nabla\times\mathbf{E}(\mathbf{r})=\mathbf{0}\ . \tag{A2}$$

The set of these two equations now replaces (1.1) and (2.9) and, hence, one expects to be able to apply a similar procedure for ω_e as for ϵ_e. From (2.9) one can see that $\mathbf{D}(\mathbf{r})$ is derivable from a vector potential

$$\mathbf{D}(\mathbf{r})=\nabla\times\mathbf{A}(\mathbf{r})\ . \tag{A3}$$

We expand $\mathbf{E}(\mathbf{r})$ in terms of $\delta\omega=\omega(\mathbf{r})-\omega_0$ and the corrections $\delta^n\mathbf{D}(\mathbf{r})$ to the uniform field $\mathbf{D}_0$, which would exist in the homogeneous medium ω_0. Requiring that (A2) is satisfied term by term, one obtains the following equation for the first variation of $\mathbf{D}(\mathbf{r})$

$$(\gamma-1)[(\nabla\delta\mathbf{D}_z)\times\hat{\mathbf{z}}]+\nabla\times\delta\mathbf{D}(\mathbf{r})=-[\nabla(\delta\omega(\mathbf{r})/\omega_0)]\times\hat{\mathbf{z}}\ . \tag{A4}$$

Rescaling only the coordinates, as done in Sec. II, i.e., $x\to x$, $y\to y$, and $z\to\zeta=\sqrt{\gamma}z$ is now insufficient for transforming (A2) into a convenient form. We also need to rescale the field $\delta\mathbf{A}$ by defining a new vector potential.

$$\mathbf{B}\equiv(\delta A_x,\delta A_y,\delta A_z/\sqrt{\gamma})\ . \tag{A5}$$

Incorporating these transformations into (A4) now yields

$$\nabla\times[\nabla\times\mathbf{B}(\mathbf{r})]=\nabla(\delta\omega(\mathbf{r})\hat{\boldsymbol{\xi}})/(\gamma\omega_0) \tag{A6}$$

and

$$\delta D_z=[\nabla\times\mathbf{B}(\mathbf{r})]_\zeta\ . \tag{A7}$$

Note that the rescaling of δA_z does not affect (A7), since δD_z depends only on δA_x and δA_y. The solution of (A6) for $\mathbf{B}$ can be formulated in terms of a tensorial Green's function $\mathcal{G}$ that satisfies

$$\nabla\times(\nabla\times\mathcal{G})-k^2\mathcal{G}=-K^2\delta^d(\boldsymbol{\rho}-\boldsymbol{\rho}')\mathcal{I}\qquad(\mathcal{G}=0\text{ on the boundaries})\ , \tag{A8}$$

where $\mathcal{I}$ is the unit tensor. The Greens's function relevant to our problem is obtained by taking the limits $k\to 0$ and $K\to 1$ at the end of the calculation.[11] Applying a Fourier transformation to both sides of (A8), one can solve for the Fourier transform of $\mathcal{G}$, $\mathcal{T}(\mathbf{q})$, to find

$$\mathcal{T}_{ab}(\mathbf{q})=\frac{K^2}{k^2}\,\frac{k^2\delta_{ab}-q_aq_b}{k^2-q^2}\ , \tag{A9}$$

where $\mathcal{T}_{ab}$ is the ab component of the tensor $\mathcal{T}$ and δ_{ab} is the Kronecker's δ function. Transforming back to coordinate space one obtains

$$\mathcal{G}_{ab}=-\frac{K^2}{k^2}\left[k^2\delta_{ab}+\frac{\partial^2}{\partial\rho_a\partial\rho_b}\right]\frac{e^{ik|\rho-\rho'|}}{4\pi|\boldsymbol{\rho}-\boldsymbol{\rho}'|}\ . \tag{A10}$$

Thus the formal solution for $\mathbf{B}$ is

$$\mathbf{B}(\mathbf{r})=\frac{1}{\gamma\omega_0}\int \mathcal{G}(\boldsymbol{\rho},\boldsymbol{\rho}')\cdot\{\nabla'\times[\delta\omega(\boldsymbol{\rho}')\hat{\boldsymbol{\xi}}']\}d^d\rho' , \tag{A11}$$

where the integration is taken in the rescaled coordinates. Letting $K\to 1$ and $k\to 0$, the surviving part of the Green's function in (A11) is

$$\mathcal{G}_{ab}=\frac{1}{4\pi|\boldsymbol{\rho}-\boldsymbol{\rho}'|} . \tag{A12}$$

Integrating (A11) by parts, discarding the surface term, and using the symmetry of $\mathcal{G}$, we find

$$\mathbf{B}(\mathbf{r})=\frac{1}{\gamma\omega_0}\int[\delta\omega(\boldsymbol{\rho}')-\langle\delta\omega\rangle]\nabla'[\mathcal{G}(\boldsymbol{\rho},\boldsymbol{\rho}')\hat{\boldsymbol{\xi}}']d^d\rho' , \tag{A13}$$

where the average $\langle\delta\omega\rangle$ has been inserted for later convenience and contributes nothing to the integral due to the vanishing of $\mathcal{G}$ on the boundary.

Next, let us define the bulk effective value of ω, via the energy density

$$\omega_e=\frac{1}{V}\int\delta\omega(\mathbf{r})\left[\frac{|\nabla\times\mathbf{A}(\mathbf{r})}{D_0}\right]^{\gamma+1}d^dr . \tag{A14}$$

As for ϵ_e, consider a variation $\delta\omega(\mathbf{r})$ and apply the δ operator to both sides of (A14) to obtain

$$\delta\omega_e=\frac{1}{V}\int\delta\omega(\mathbf{r})\left[\frac{|\nabla\times\mathbf{A}_0|}{D_0}\right]^{\gamma+1}d^dr=\langle\omega\rangle-\omega_0 . \tag{A15}$$

The term within the integrand, that contains the first variation of $|\nabla\times\mathbf{A}|^{\gamma+1}$, vanishes as a consequence of the variational property of (3.27). The second-order correction to ω_e is

$$\delta^2\omega_e=\frac{1}{V}\int\delta\omega\,\delta\left[\frac{|\nabla\times\mathbf{A}|}{D_0}\right]^{\gamma+1}d^dr ,$$

or, after some simplifications and transformation to the rescaled coordinates,

$$\delta^2\omega_e+\frac{\gamma+1}{\sqrt{\gamma}V}\int d^d\rho[\delta\omega(\boldsymbol{\rho})-\langle\omega\rangle][\nabla\times\delta\mathbf{A}(\mathbf{r})]_\xi . \tag{A16}$$

The insertion of $\langle\delta\omega\rangle$ into the integrand contributes nothing due to the boundary conditions on $\delta\mathbf{A}$. Using (A5) and (A13) to substitute for $\delta\mathbf{A}$ in (A16), we get

$$\delta^2\omega_e=\frac{\gamma+1}{\gamma^{3/2}\omega_0V}\int d^d\rho\int d^d\rho'[(\delta\omega(\boldsymbol{\rho})-\langle\omega\rangle)\times\delta\omega(\boldsymbol{\rho}')-\langle\omega\rangle]\times[\nabla\times(\epsilon_{\xi cb}\partial'_c\mathcal{G}_{ba}\hat{\boldsymbol{\xi}})]_\xi , \tag{A17}$$

where, for brevity, we have adopted the convection of summation over repeated indices, and where $\epsilon_{ijk}=1,-1$, and 0 if i,j, and k are cyclic, anticyclic, or when two of them are equal, respectively. Next we replace the term in the square brackets by its average, the correlation function

$$f(\boldsymbol{\rho},\boldsymbol{\rho}')=\langle[\delta\omega(\boldsymbol{\rho})-\langle\delta\omega\rangle][\delta\omega(\boldsymbol{\rho}')-\delta\omega\rangle]\rangle . \tag{A18}$$

The conditions that permit this are the same as those justifying the analogous procedure in the case of ϵ_e, namely, the volume of integration must be larger than the microgeometric correlation length and the medium must be macroscopically homogeneous.

Due to the macroscopic homogeneity, f is a function only of the relative coordinate $\mathbf{R}$. Under the conditions stated above, and far from the boundaries, the exact Green's function may be replaced by the one in (A12) (which corresponds to an infinite volume). With this (A17) becomes

$$\delta^2\omega_e-\frac{\gamma+1}{\gamma^{3/2}\omega_0V}\int d^d\rho\int d^d\rho' f(\mathbf{R})(\partial_{cc}\mathcal{G}_{bb})(\epsilon_{\xi cb})^2 , \tag{A19}$$

where the summation is over $b,c=x,y$. To make further progress we consider the Fourier transforms $\mathcal{F}_{ab}(\mathbf{q})$ and $\tilde{f}(\mathbf{k})$ of $\mathcal{G}_{ab}(\mathbf{R})$ and $f(\mathbf{R})$, respectively. The correlation function in the original system f_0 depends on $|\mathbf{R}|$ alone. It follows that its Fourier transform

$$\tilde{f}_0(\mathbf{k})=\int e^{i\mathbf{k}\cdot\mathbf{R}}f_0(\mathbf{R})d^dR ,$$

is a function only of $|\mathbf{k}|$. Calculating the transform in the rescaled coordinates, one finds

$$\tilde{f}(\mathbf{k})=\sqrt{\gamma}\int f_0(\mathbf{R})\exp[i(k_xx+k_yy+\sqrt{\gamma}k_zz)]d^dk , \tag{A20}$$

and hence

$$\tilde{f}(\mathbf{k})=\sqrt{\gamma}\tilde{f}_0\{|k^2[1+(\gamma-1)\cos^2\theta]|\} , \tag{A21}$$

with θ being the polar angle between $\mathbf{k}$ and the ξ axis. The Fourier transform of the Green's function, given in (A9), can be used to yield

$$\partial_{cc}\mathcal{F}_{bb}(\mathbf{q})=\frac{q_x^2+q_y^2}{|q^2|}=1-u^2,\quad u\equiv q_\xi/|q| . \tag{A22}$$

Now it is possible to rewrite (A19) in three dimensions as

$$\delta^2\omega_e=-\frac{\gamma+1}{\gamma\omega_0V}\int d^3\rho\int d^3R(1-u^2)\tilde{f}_0\{|k^2[1+(\gamma-1)u^2]|\}\frac{d^3k}{(2\pi)^3}\frac{d^3q}{(2\pi)^3}e^{-i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}} . \tag{A23}$$

The integration over $\mathbf{R}$, ρ, and $\mathbf{q}$ can be carried out immediately, and the remaining integral over $\mathbf{k}$ can be simplified in polar coordinates by changing the radial variable to $K \equiv [1+(\gamma-1)u^2]^{1/2}k$. This yields

$$\delta^2\omega_e = -\frac{\gamma+1}{2\omega_0\sqrt{\gamma}} \int \tilde{f}_0(|K|)\frac{4\pi K^2}{(2\pi)^3}dK \times \int_{-1}^{1} \frac{(1-u^2)}{[1+(\gamma-1)u^2]^{3/2}}du \ . \quad \text{(A24)}$$

The integral over K can be identified as $f_0(\mathbf{R}=\mathbf{0})$, while the integral over u can be carried out explicitly to yield in the final analysis

$$\delta^2\omega_e = -\frac{1+\gamma}{1-\gamma}\frac{f_0(0)}{\omega_0}\left[\frac{\arcsin(\sqrt{1-\gamma}}{\sqrt{\gamma(1-\gamma)}}-1\right] . \quad \text{(A25)}$$

The generalization of this result to higher dimensions d is straightforward and only requires the replacement of the power $\frac{3}{2}$ in the last integral over u, in (A24), by $d/2$.

Finally this second-order contribution can be added to the zero and first orders to yield an expression for ω_e that is correct to second order in $\delta\omega$,

$$\omega_e = \langle\omega\rangle - \frac{\beta+2}{2\beta}\left[\frac{\beta+1}{\sqrt{\beta}}\arcsin[\sqrt{\beta/(\beta+1)}]-1\right] \times \langle\delta\omega^2\rangle/\omega_0 + O((\delta\omega)^3) \ , \quad \text{(A26)}$$

where we have substituted $\gamma = 1/(\beta+1)$ for comparison with the result for ϵ_e. This comparison indeed shows that when ω_e is expanded in terms of $\delta\epsilon$ rather than $\delta\omega$, the two expressions are in agreement to $O((\delta\epsilon)^2)$. This is expected, of course (see the discussion in Sec. III concerning the compatibility of the definitions of ω_e and ϵ_e).

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